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A 4000 Mc/s WIDE-BAND AMPLIFIER USING A DISC-SEAL TRIODE

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Although in recent years several special types of electronic tubes, such as travelling-wave tubes and klystrons, have been developed for use as amplifiers at very high frequencies, the oldest type of amplifying tube, the triode, can still be put to good use for this purpose. Following upon an earlier article, which described the design of a disc-seal triode, type EC 57, capable of operating at frequencies up to 4000 Mc/s, the present article describes an amplifier in which this tube can be employed. The amplifier is now being used, to give one example, for the microwave telecommunication network at present in course of construction in the Netherlands.

Introduction

In recent years work has been going on in many parts of the world on the construction of microwave radio links, intended for relaying multichannel telephony and television signals. A frequency band from 3800 to 4200 Mc/s (wavelength approx. $7\frac{1}{2}$ cm) has been reserved for these links by international agreement. These frequencies, while being high enough to allow a satisfactory directional effect to be obtained without the use of too large reflectors, are not so high as to give rise to excessive fading in unfavorable weather conditions. In the present article a description will be given of an amplifier specially developed for the frequency range in question. The amplifier, which is at present being employed in the microwave telecommunication network in course of construction in the Netherlands, can be operated with two types of disc-seal triode, viz. type EC 56 and EC 57. With the EC 57 tube, which has already been discussed in this Review ¹⁾, the amplifier can deliver a power of about 1.5 W. Although this is lower than the output at the same frequency of the multi-reflex klystron, also described on an earlier occasion ²⁾, the amplifier dealt

with here will nevertheless be an important asset in fields of application for which the multi-reflex klystron is unsuitable.

The multi-reflex klystron can only be employed as an oscillator, for which reason it is used, for example, in beam transmitters with double frequency modulation ³⁾. Owing to the relatively low frequency of the auxiliary carrier wave, however, this system cannot be used when it is necessary to transmit signals with a very wide frequency band, as in television. In that case, only single modulation is possible, and when, as is usual in relay and terminal stations, the amplification is obtained at an intermediate frequency much lower than that of the signals to be transmitted, a mixer stage must be used to achieve the latter frequency. At centimetric waves a crystal diode is suitable for this purpose. Its output, however, being only a few milliwatts, is inadequate for establishing a reliable radio link. An amplifier is then needed which operates at the transmitter frequency.

Microwave amplifiers

Before describing the design of an amplifier for centimetric waves, we shall deal briefly with the main differences between amplifiers of this kind and those which operate at lower frequencies.

At frequencies above 500 Mc/s it is almost impossible to build resonant circuits composed of coils and capacitors. It is then necessary to resort to Lecher systems and, at frequencies above about

¹⁾ G. Diemer, K. Rodenhuis and J. G. van Wijngaarden, The EC 57, a disc-seal microwave triode with L cathode, Philips tech. Rev. **18**, 317-324, 1956/57 (No. 11).

²⁾ F. Coeterier, The multireflection tube, a new oscillator for very short waves, Philips tech. Rev. **8**, 257-266, 1946.
F. Coeterier, The multi-reflex klystron as a transmitting valve in beam transmitters, Philips tech. Rev. **17**, 328-333, 1955/56.

³⁾ See C. Ducot, Beam transmitters with double frequency modulation, Philips tech. Rev. **17**, 317-327, 1955/56.

2000 Mc/s, to resonant cavities. In the latter case the various stages of an amplifier are usually coupled by waveguides, and this fact must be taken into account in the design of the amplifier.

When using a triode in the centimetric wave range the only possible circuit is the grounded grid arrangement. The fairly low input impedance of the tube in this circuit entails a rather high driving power; this power has to be fed to the tube input via a waveguide. It is desirable that the tube should then be, as far as possible, a matched load to the waveguide. With centimetric waves it is hardly possible to use anything but a resonant cavity as the anode circuit, and this must be coupled to the output waveguide. The circuit must be designed such that the tube can provide the desired gain at the operating frequency, with a sufficiently large bandwidth. Moreover, the anode resonant cavity must be tunable. We shall deal in this article with various problems arising in this connection.

Construction of the amplifier

Fig. 1 shows in schematic form the construction of the amplifier⁴⁾. It consists of a block of brass into which slots have been cut to form the input and output waveguides, *E* and *B* respectively. The two waveguides are coupled by a cylindrical hole *D*. The tube is inserted from underneath through the input waveguide, the grid disc *G* being screwed in the upper wall of *E*. The cathode disc *F* is insulated for DC from the underside of the input waveguide. For the operational frequency range, however, the coupling between these elements is practically a short-circuit⁵⁾. The cathode base *K* protrudes through the input waveguide, which is short-circuited at a distance *a* of about one-quarter wavelength from the centre of the tube.

In the cylindrical cavity *D* there is an inner conductor *H*, the top of which is supported by a disc *J* of a synthetic material (polytetrafluorethylene) while the bottom makes spring contact with the anode disc. The cavity *D* also contains a plunger *Z*, which is in contact with the wall of *D* but not with the inner conductor *H*. The plunger thus forms with the part of *H* which it covers a coaxial line whose length *b* is approximately one-quarter

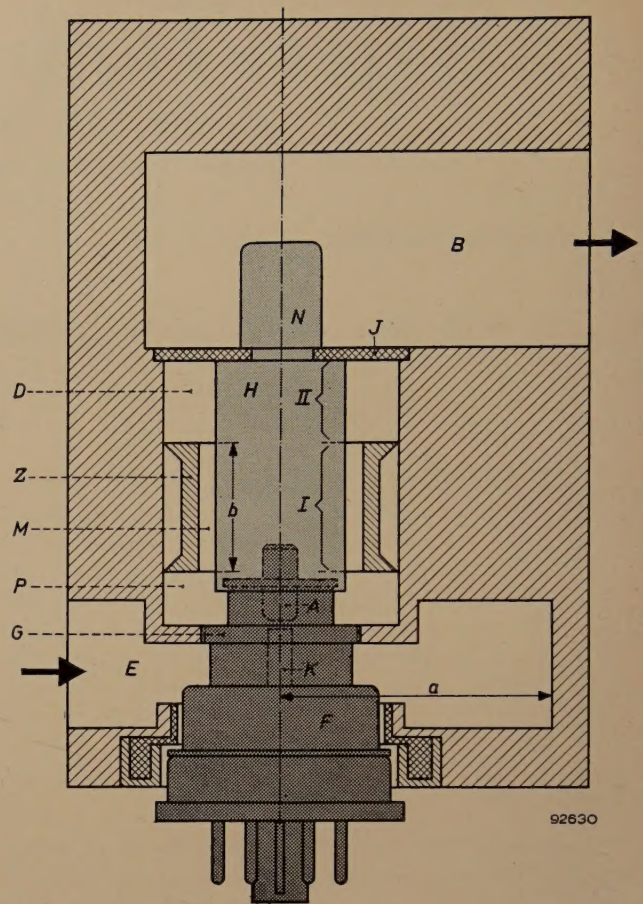


Fig. 1. Simplified cross-section of the amplifier. This consists of a block of brass, in which slots are cut to form the input and output waveguides, *E* and *B* respectively. Between these waveguides there is a cylindrical hole *D*. *G*, *F*, *K* and *A* are respectively the grid disc, the cathode disc, the cathode base and the anode of the disc-seal triode (EC 56 or EC 57). In cavity *D* there is an inner conductor *H*, which is supported by a disc *J* of plastic and which, with an extension piece *N*, protrudes into waveguide *B*. The anode resonant cavity *P* is tuned by a plunger *Z*. *M* is the gap between *H* and *Z*. *H*, *Z* and the wall of cavity *D* form two coaxial lines, denoted by *I* and *II*. The arrows indicate the direction in which the input power enters and the output power leaves the amplifier.

of a wavelength. We shall refer to this coaxial line as *I*. Since the gap *M* between *Z* and *H* is fairly narrow, the characteristic impedance of *I* is low.

The section of the inner conductor above the plunger forms, with the wall of *D*, another coaxial line, which we shall refer to as *II*. Its characteristic impedance is higher than that of *I*. The inner conductor *H* has an extension piece *N* which protrudes into the output waveguide *B*. The dimensions of this section are so chosen as to make its radiation resistance over a wide frequency band equal to the characteristic impedance of coaxial line *II*. If waveguide *B* has a matched termination, the same will apply to the top end of coaxial line *II*.

At the bottom end of the plunger a resonant cavity *P* is formed, which incorporates the grid-anode capacitance of the tube. This cavity is inter-

⁴⁾ A similar amplifier has already been described by A. E. Bowen and W. W. Mumford, A new microwave triode: its performance as a modulator and as an amplifier, Bell Syst. tech. J. 29, 531-552, 1950.

⁵⁾ A coupling of this kind is known as a "choke coupling"; see e.g. G. L. Ragan, Microwave transmission circuits, Radiation Laboratory Series, No. 9, McGraw-Hill, New York 1948.

rupted at the top by the bottom of gap M . Coaxial line I now functions as a quarter-wavelength transformer, the top end of which is terminated by the characteristic impedance of coaxial line II . The input impedance at the bottom end of this transformer is therefore very low (approximately 0.5Ω), and it is this that forms the load of the anode resonant cavity P . The cavity is tuned by moving the plunger Z . (Owing to the matched termination of coaxial line II , the tuning does not alter the load resistance of the anode resonant cavity.)

The anode voltage is fed to the tube via a thin wire (see Y in fig. 13) which runs from N to the wall of the waveguide as far as possible at right angles to the electrical lines of force; it is led out via a disc capacitor mounted in a coaxial plug.

The input circuit

At a frequency of 4000 Mc/s the input impedance of the tube in the amplifier is almost a pure resistance, the value of which roughly corresponds to that needed for the waveguide E to be matched.

This matched termination gives rise to a standing-wave ratio of 1 in the waveguide. With a given tube, however, this is not readily possible for all frequencies in the desired frequency band. By way of example, fig. 2 shows the standing-wave ratio in the waveguide E plotted as a function of the frequency. It can be seen from this figure that the input of the tube behaves rather like a tuned circuit.

At low frequencies the input resistance of a triode with grounded grid is approximately equal to the reciprocal value of the transconductance. In the case of the EC 57, the transconductance is approximately 20 mA/V, and hence the input resistance should be 50Ω . As explained in the article quoted under ¹⁾ the transconductance at 4000 Mc/s is about 30% lower than at low frequency, in which case therefore the input resistance should be 70Ω . It must be taken into account that this input resistance is directly between grid and cathode.

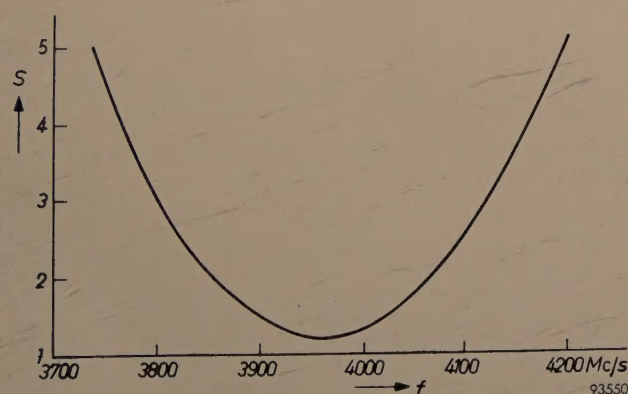


Fig. 2. Standing-wave ratio S in the input waveguide as a function of the frequency f .

At the high frequencies in question the equivalent resistance of the tube between cathode and grid disc has a different value, owing to the influence of the self-inductances and mutual capacitances of the electrodes. An equivalent circuit for the input impedance of the tube is shown in fig. 3. Here, R_{gk} is the input resistance, measured directly between grid and cathode and C_{gk} is the capacitance between these electrodes, while L_k is the inductance of the cathode base and the cathode disc. In parallel to these is the capacitance C_d between grid disc and cathode disc. A simple calculation reveals that at a frequency of 4000 Mc/s the equivalent resistance of this circuit is approximately a pure resistance of 400Ω , which more or less corresponds to the characteristic impedance of the waveguide.

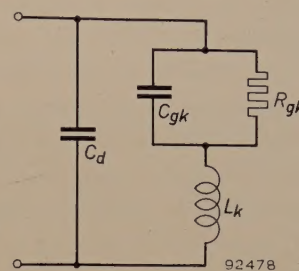


Fig. 3. Equivalent circuit of the amplifier input. R_{gk} is the input resistance between grid and cathode, C_{gk} the capacitance between these electrodes; L_k the inductance of cathode base and cathode disc, and C_d the capacitance between grid disc and cathode disc.

In order to be able, with individual tubes, to match the input end of the tube to the waveguide at any desired frequency in the frequency band to be covered, variable reactive elements have been introduced at two different points in the waveguide; these are not shown in fig. 1, but can be seen separately in fig. 4a. Each of these elements consists of a screw S in the centre of the upper wall of the waveguide, and a rod T joining the upper and lower walls, away from the centre. Waveguide E contains two such combinations at a distance d of approximately one-eighth of a wavelength apart.

It can be shown with the aid of the theory of electromagnetic wave propagation in waveguides that a rod, as represented by T , has the same effect on these waves as a coil, mounted between the two conductors of a Lecher line, would have on wave propagation along the latter. The inductance of this equivalent coil depends upon the position and the thickness of the rod. On the other hand, a screw S (fig. 4a) is equivalent to a capacitor connected between the conductors of a Lecher line. The capacitance of S depends upon the depth to which it protrudes into the waveguide. The combination of S and T is comparable, then, with a resonant circuit between the conductors of a Lecher line. Fig. 4b shows an electrically equivalent circuit, in which the waveguide is replaced by a Lecher line. The two capacitors (i.e. the screws S) can be so adjusted as to make the circuits either capacitive or inductive, and it is possible in this way, with varying tube impedances and with different frequencies, to match the waveguide E perfectly.

Although the two reactive elements make it possible to match the tube to the input waveguide E under all practical conditions, they nevertheless reduce the bandwidth in which a sufficiently small

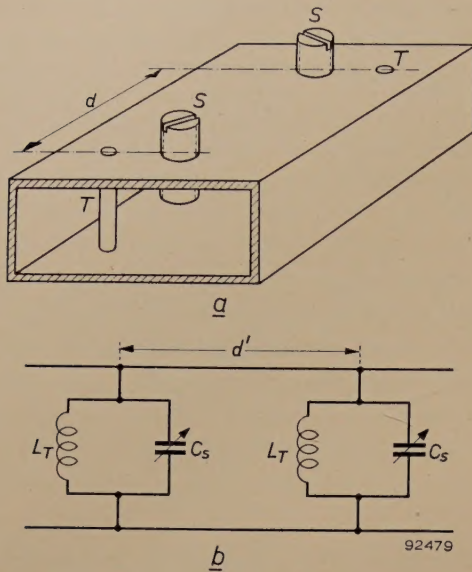


Fig. 4. a) The input waveguide contains two variable reactive elements, each of which consists of a screw S and a rod T . The distance d is approximately one-eighth wavelength. b) Equivalent circuit of the arrangement in fig. 4a. C_S and L_T are respectively the "capacitance" of the screw S and the "inductance" of the rods T . The distance d' is about one-eighth wavelength.

reflection appears at each side of the tuned frequency. This bandwidth, however, is still much larger than that of the anode circuit, and therefore the latter mainly determines the total bandwidth of the amplifier.

The anode circuit

In order to make the frequency characteristic flat enough over a wide frequency band, substantial damping of the resonant cavity P (see fig. 1) would be necessary. This would reduce the resonance resistance to the extent that no gain would be obtained. A familiar method of obtaining a sufficiently high gain over a wide frequency band at lower frequencies is to employ a band-pass filter, consisting of two resonant coupled circuits. It would be rather difficult mechanically to arrange a second resonant cavity near to P and directly coupled with it, particularly since both cavities would have to be tunable. In very-high frequency technique, however, it is possible to couple two resonant circuits at some distance apart by means of a suitable length of transmission line, which may consist of a coaxial line, a Lecher

line, a waveguide or a combination of these. The latter is used in the present case. The second resonant circuit is contained in waveguide B , and consists of an inductance formed by two rods T_i joining the upper and lower walls, and a capacitance formed by a screw S_i through the upper wall (see fig. 5). This circuit, known as a resonant iris, will be referred to in the following simply as "iris"⁶⁾. The position of this iris is so chosen as to make the system between the top of the plunger S and the iris electrically equivalent to a line one quarter-wave length long.

An equivalent circuit for the whole anode system is given in fig. 6a, in which C_a and L_a represent the resonant cavity P . The tube is represented by a voltage source V_a in series with the circuit L_a , C_a . (The damping effect of the internal resistance of the tube on the resonant cavity can be disregarded here.) The resonant circuit formed by L_i and C_i represents the iris sketched in fig. 5. This circuit is damped by the characteristic impedance R_0 of waveguide B . The coupling between both circuits, which in reality consists of the coaxial line I (see fig. 1), the coaxial line II , the extension piece N of the inner conductor H and the output waveguide B , is represented in fig. 6a by a transformer Tr having a transformation ratio n ⁷⁾. We shall now proceed, with reference to this diagram, to calculate the voltage V_i when the frequency of V_a is varied. For this purpose we shall substitute the section of the circuit on the left of L_i , C_i by a voltage source $V_a' = V_a/n$ in series with a capacitor having a capacitance $C_a' = n^2 C_a$, and a coil having an inductance $L_a' = L_a/n^2$ (fig. 6b). The circuit L_a' , C_a' is

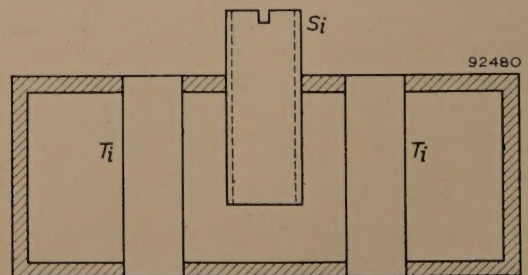


Fig. 5. The output waveguide contains a resonant iris, which consists of a screw S_i and two rods T_i .

- ⁶⁾ The reason is that a reflecting element of this kind placed in a waveguide has the same effect as a conductive plate with a hole in it; see e.g. C. G. Montgomery, R. H. Dicke and E. M. Purcell, *Principles of microwave circuits*, Radiation Laboratory Series, No. 8, McGraw-Hill, New York 1948, Chapter 6.
- ⁷⁾ This transformation ratio is dependent on the frequency; it appears, however, that sufficiently accurate results are obtained from a calculation in which n in the frequency band concerned is regarded as constant.

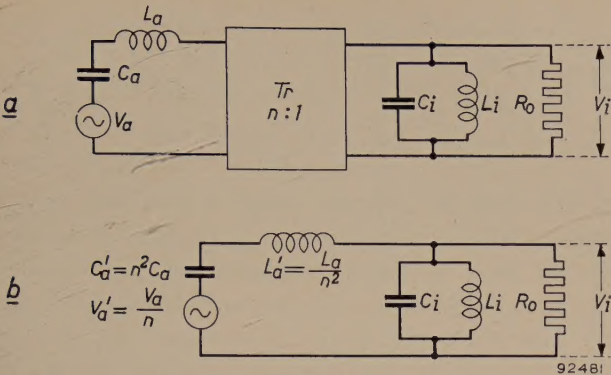


Fig. 6. a) Equivalent circuit in which the tube is represented by a voltage source V_a . L_a and C_a represent the anode resonant cavity, while L_i and C_i represent the iris. The iris is damped by the characteristic impedance R_0 of the output waveguide. The transmission system between resonant cavity and iris is represented by a transformer Tr with a transformation ratio n .
b) The section on the left of L_i , C_i is replaced by a voltage source V_a' in series with a capacitor C_a' and a coil L_a' .

then tuned to the same frequency as the circuit L_a , C_a , and hence the same frequency as L_i , C_i . The relevant angular frequency will be denoted ω_0 . We shall also introduce the following quantities:

$Q_a = \frac{\omega_0 L_a'}{R_0}$, the figure of merit of circuit L_a' , C_a' if the circuit L_i , C_i is not present. Evidently, this is also the figure of merit of the anode resonant cavity without the iris.

$Q_i = \frac{R_0}{\omega_0 L_i}$, the figure of merit of circuit L_i , C_i if this is damped by R_0 .

$k = \frac{Q_i}{Q_a}$, the coupling factor, the meaning of which will be discussed later.

$\beta = \frac{\omega}{\omega_0} - \frac{\omega_0}{\omega}$. If the difference $\Delta\omega$ between ω and ω_0 is small with respect to ω_0 , we may write $\beta = 2\Delta\omega/\omega_0$.

For the ratio between the voltages V_i and V_a' the following equation may now be derived:

$$\frac{V_i}{V_a'} = \sqrt{1 + (\beta Q_a)^2 (1 - 2k) + (\beta Q_a)^4 k^2}^{-1/2}. \tag{1}$$

In fig. 7, V_i/V_a' is plotted as a function of βQ_a for several values of k . For large values of k the curves show two peaks, for small values only one. The transition between these two situations takes place when

$$k = \frac{1}{2}. \tag{2}$$

The quantity Q_a is dependent on the transformation ratio n and hence of the strength of the coupling between the resonant cavity and the rest of the circuit. A small value of n is associated with a large value of L_a' , and thus also with a large value of Q_a . In this case, then, k is small, and the resonant cavity is loosely coupled to the remaining part of the anode circuit. For this reason the quantity k is termed the coupling factor. The situation where $k = \frac{1}{2}$ is known as “transitional coupling”⁸⁾.

As appears from fig. 7, the curve is flattest at approximately the value of k given by (2); it is therefore desirable to design the anode circuit so as to approximate to the transitional coupling. According to the definition of k , the figure of merit of the iris should be half that of the anode resonant cavity.

8) See e.g. C. B. Aiken, Two-mesh tuned coupled circuit filters, Proc. Inst. Rad. Engrs. **25**, 230-272, 1937. This term should not be confused with the term critical coupling which, with a band-pass filter both of whose circuits are equally damped, designates the coupling which gives maximum gain and for which the resonance curve is only just short of having two peaks. Although the transitional coupling in the circuit of fig. 6b corresponds to the latter situation, in this case the maximum value of V_i/V_a' does not occur. For $\beta = 0$ it follows from formula (1) that $V_i/V_a' \equiv n V_i/V_a = 1$. We see, then, that V_i/V_a' is independent of n , while V_i/V_a is inversely proportional to n .

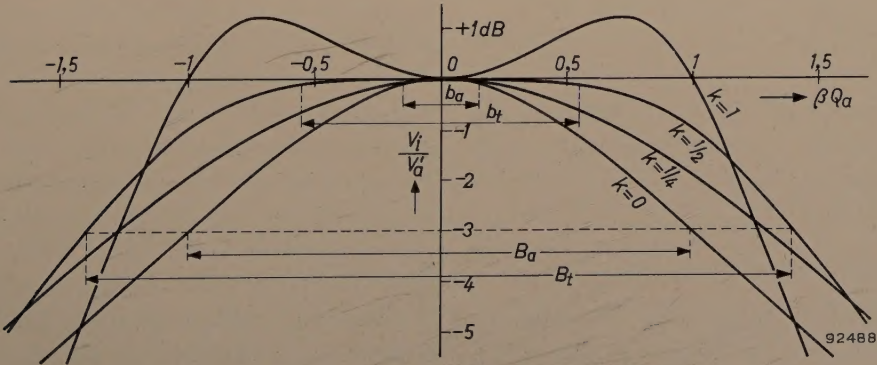


Fig. 7. The voltage ratio V_i/V_a' from fig. 6b, plotted in dB as a function of the product βQ_a . The curves hold for three different values of k . At $k = \frac{1}{2}$, transitional coupling is obtained.

The curve for $k = 0$ in fig. 7 indicates the value of V_i/V_a' for the case where no iris is present ($Q_i = 0$ means $L_i = \infty$, hence $C_i = 0$). This, then, is the resonance curve of the anode cavity separately.

The usual method of measuring the figure of merit of tuned circuits is to determine the width of the frequency band at the limits of which the circuit impedance is a factor $\sqrt{2}$ smaller than the resonance resistance. At the edges of this frequency band, the response is 3 dB down. This bandwidth of the resonant cavity is denoted in fig. 7 by B_a . If the corresponding bandwidth of the iris be denoted by B_i , it follows from the fact that the bandwidth of the circuit is inversely proportional to the figure of merit that:

$$B_i = 2B_a. \quad \dots \dots (3)$$

In fig. 7, B_t indicates the width of the frequency band at the edges of which the curve for $k = \frac{1}{2}$ has fallen to 3 dB below peak value. It is seen that $B_t = \sqrt{2}B_a$. The improvement obtained by applying the iris is, however, much larger than this factor $\sqrt{2}$. The reason is that in order to keep the distortion of the transmitted signals within reasonable limits, a much smaller frequency band can be used than is indicated by B_t in fig. 7. As a rule, the requirement is that the amplification at the edges of the frequency band should drop by no more than 0.1 dB. In the following we shall denote this bandwidth by a small letter b in order to distinguish it from the earlier-mentioned bandwidth B . It will be clear from a consideration of fig. 7 that in this respect the curve for $k = \frac{1}{2}$ is appreciably better than that for $k = 0$. For a single tuned circuit the bandwidth b_a in question is

$$b_a = 0.15 B_a.$$

If transitional coupling is now introduced between this circuit and a second, whose bandwidth is $2B_a$, the resultant curve will fall to 0.1 dB below the peak value at a width b_t which is given by

$$b_t = 0.39 B_t = 0.55 B_a.$$

Thus, by introducing the iris, the width of the frequency band at the edges of which the amplification has dropped by only 0.1 dB is increased by a factor $0.55/0.15 = 3.7$.

Adjustment to the required bandwidth

When the frequency band b_t of an amplifier constructed according to the foregoing principle is required to have a specific value, it is easy to calculate what values of the bandwidths B_a and B_i

of the anode resonant cavity and the iris are required, since

$$B_a = \frac{1}{0.55} b_t = 1.82 b_t \text{ and } B_i = 2 B_a = 3.64 b_t.$$

If, for example, constant amplification is required over a frequency band of 55 Mc/s, the bandwidth B_a of the amplifier without iris should be $1.82 \times 55 = 100$ Mc/s and the bandwidth B_i of the iris $3.64 \times 55 = 200$ Mc/s. In order to adjust the latter band width to the required values an experiment is needed to determine the position and thickness of the rods T_i (see fig. 5).

The iris is placed in a waveguide, one end of which is matched and the other end connected to an energy source, also matched to the waveguide. The iris is then damped, both by the load and by the energy source, with an impedance equal to the characteristic impedance of the waveguide. An equivalent circuit, in which the impedances are represented as lumped circuit elements, is shown in fig. 8. The circuit L_i, C_i is now damped by a resistance $\frac{1}{2}R_0$, and it is thus evident that the circuit must be so designed that its bandwidth in this arrangement will be equal to $2B_i = 7.3 b_t$, in our case 400 Mc/s. This situation can be achieved by a suitable choice of the position and thickness of the rods T_i (fig. 5).

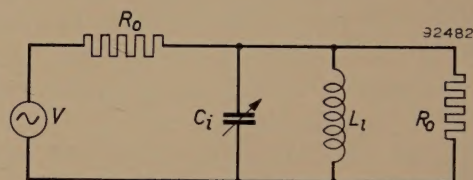


Fig. 8. Equivalent circuit for the iris, placed in a waveguide with matched termination; the waveguide is connected to an energy source which is also matched.

Once the rods have been placed in the amplifier with the correct thickness and at the correct place, the resonant cavity and iris must be tuned to the required central frequency, and the coupling adjusted such that $k = \frac{1}{2}$. The resonant cavity is tuned with the plunger Z (fig. 1) and the iris with the screw S_i (fig. 5). To adjust the coupling it is necessary to vary the figure of merit Q_a of the resonant cavity. This is done by changing the transformation ratio n (see fig. 6). One way might be to vary the characteristic impedance of the quarter-wavelength transformer by altering the radial dimensions of gap M (see fig. 1). Mechanically, this would be difficult; however, for small variations, the same effect can be achieved with a construction in which a reactive element is placed in the waveguide at a distance of one-eighth of a wavelength from the iris. This element again consists of a rod joining the upper and lower walls of the waveguide and of

a screw through the upper wall. In *fig. 9* a cross-section of the output waveguide is shown in which this reactive element is represented by a screw S'

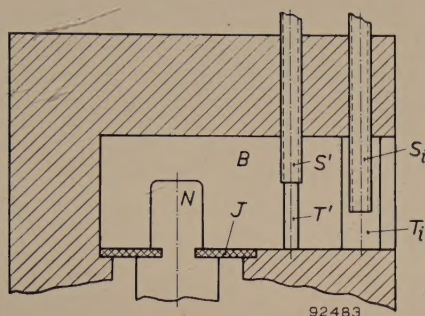


Fig. 9. Cross-section of the output guide, in which the iris is denoted by S_i , T_i , and the reactive element with which the coupling is adjusted is denoted by S' , T' . For the meaning of letters B , N and J see *fig. 1*.

and a rod T' . Now, it can be shown that the effect of screwing S' in and out is to vary the transformation ratio n of the transmission system between resonant cavity and iris, and also to change V_a' , L_a' and hence Q_a . Since Q_i remains constant, the coupling factor k is also varied during this process.

Cooling

In the construction of the amplifier as shown in *fig. 1* the anode section of the tube is entirely incorporated in the inner conductor H . There is therefore a danger that the anode disc will be overheated at the maximum anode dissipation of 10 W and for this reason, measures have been taken to cool the inner conductor H . For this purpose, H could be made hollow and provided internally with radial cooling fins. *Fig. 11* shows a cross-section of such an inner conductor, in which the direction of air flow is indicated by arrows. Through a polystyrene tube U the cooling air is fed in from outside to the extension piece N of the inner conductor H . The air flows via the cooling fins V to the output waveguide B , where it leaves the amplifier. The quantity of air required is about 6 l/min at a pressure of approximately 5 cm water.

The complete amplifier

Fig. 12 is a photograph of an amplifier as seen from the input and output sides; *fig. 13* gives a perspective cut-away sketch of the complete

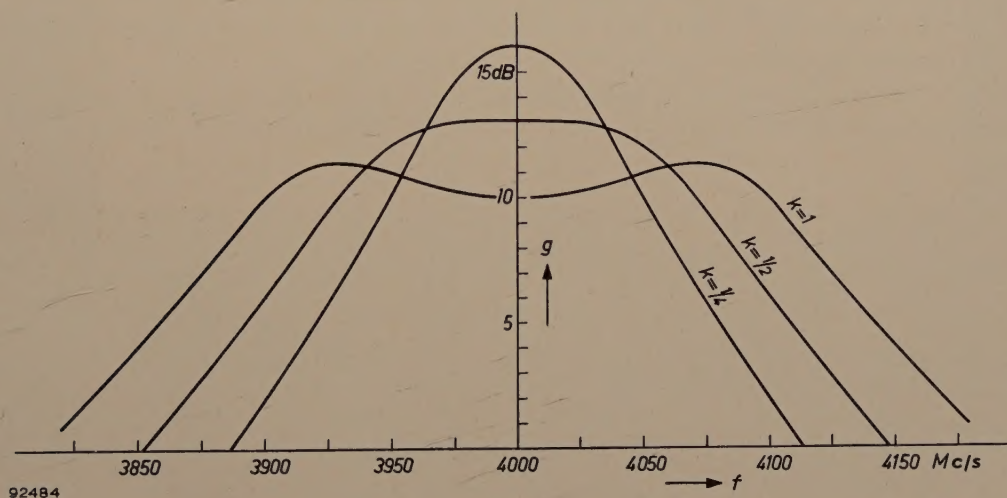


Fig. 10. The gain obtained with an tube EC 57, tuned to a central frequency of 4000 Mc/s, for various values of the coupling factor k , and an iris bandwidth B_i of 200 Mc/s.

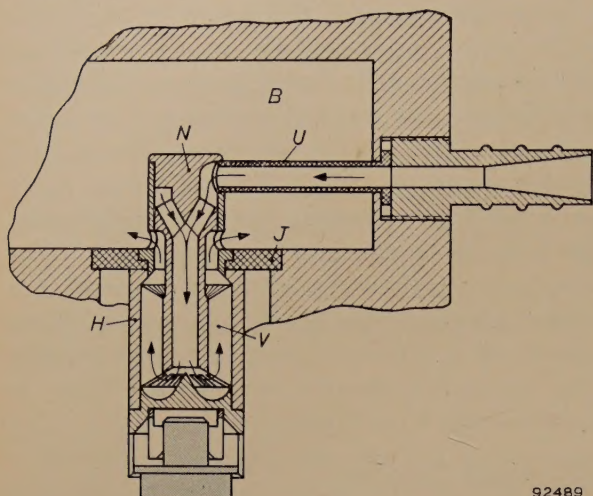
As regards the curves in *fig. 7*, this affects both the two scales and the running parameter k , so that the total effect of turning screw S' is difficult to see from this figure. A good impression of what happens when the coupling screw S' is turned is given by *fig. 10*, in which, for an EC 57 tube, the amplification in dB is plotted against the frequency for several values of k . It can be seen that as k decreases, the amplification at the central frequency increases, although the bandwidth becomes smaller. At the transitional coupling ($k = \frac{1}{2}$) the amplification at the central frequency is 13 dB.

amplifier showing the control elements discussed in the foregoing.

The plunger Z is moved by two rods W fixed at pivots to a cross-beam X , forming, as it were, a balance. In this way the plunger is always subjected to two equal forces, thereby precluding mis-alignment.

The EC 57 tube¹⁾ can be used in this amplifier. If a smaller output power is adequate, a type EC 56 tube may be used instead, the nominal anode current of which is half that of the EC 57 (30 against 60 mA). This current is set to the correct value by

means of a variable cathode resistor; to reduce the effect of deviations in the characteristics of individual tubes, the cathode resistor can be connected to a point at a negative potential.



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Fig. 11. A system for cooling the amplifier. The cooling air is fed through the polystyrene tube *U* to the cooling fins *V*. For the meaning of the letters *B*, *H*, *J* and *N* see fig. 1.

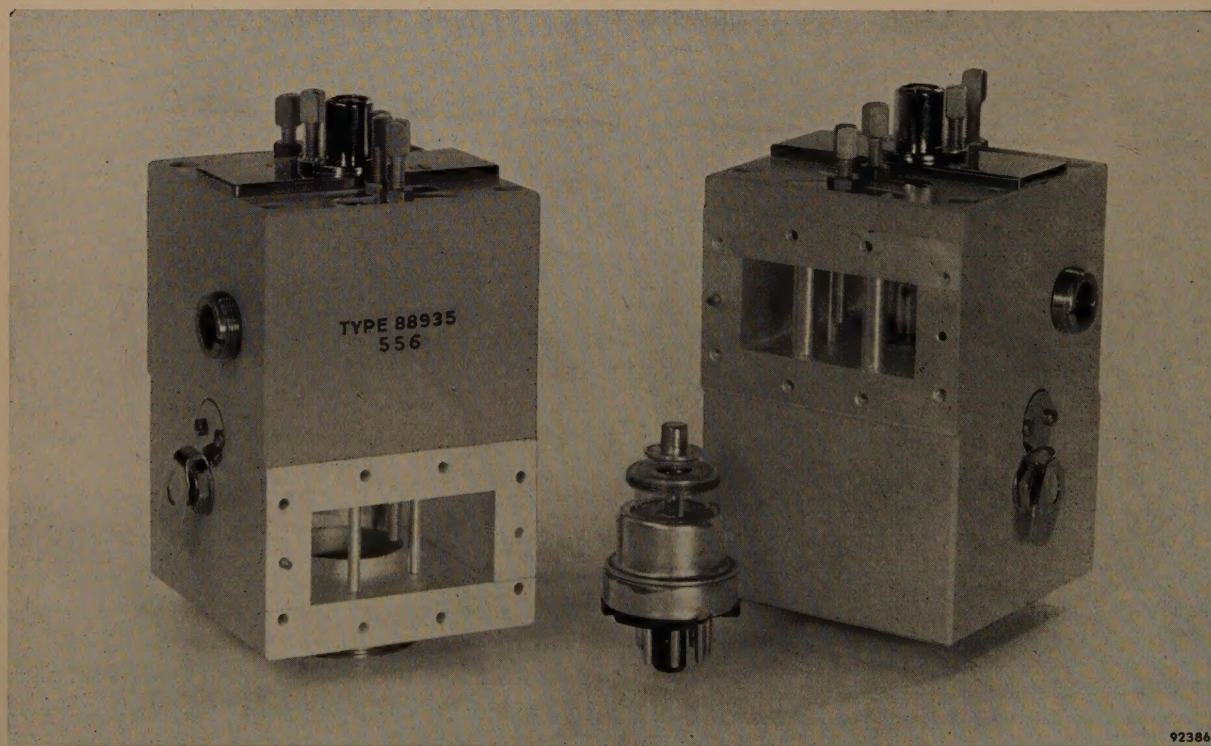
The gain obtainable in the case of small signals is 12 dB with the EC 56 and 13 dB with the EC 57 (at a bandwidth b_t of 55 Mc/s); for larger signals the gain drops. In fig. 14 the gain for both tubes is plotted as a function of the output power. It can be seen that

with a gain of 8 dB an output of 0.5 W can be obtained from the EC 56, while with the same gain the EC 57 delivers a power of 1.5 W. Fig. 14 holds for an amplifier with a bandwidth b_t adjusted to 55 Mc/s. As mentioned in the article quoted under ¹⁾, a higher gain can be obtained with a smaller bandwidth, in such a way that the product of bandwidth and gain is almost constant. The bandwidth cannot, however, be reduced too far: at a bandwidth of 10 to 20 Mc/s the amplifier becomes unstable owing to the feedback occurring in the tube.

Feedback

When the tube is in operation, feedback occurs owing to the small but by no means negligible capacitance between anode and cathode. The reason for this is that the grid does not completely screen these two electrodes from each other. A second cause of feedback is the fact that, owing to the electromagnetic alternating field in the anode resonant cavity, currents are induced in the grid wires, and these currents in their turn give rise to an alternating voltage between grid and cathode. The effect of this can be accounted for in the equivalent circuit by a coil in series with the grid connection ⁹⁾.

⁹⁾ See G. Diemer, Passive feedback admittance of disc-seal triodes, Philips Res. Rep. 5, 423-434, 1950.



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Fig. 12. Two amplifier blocks, the left one seen from the input side, the right one from the output side. The EC 57 tube is shown between them.

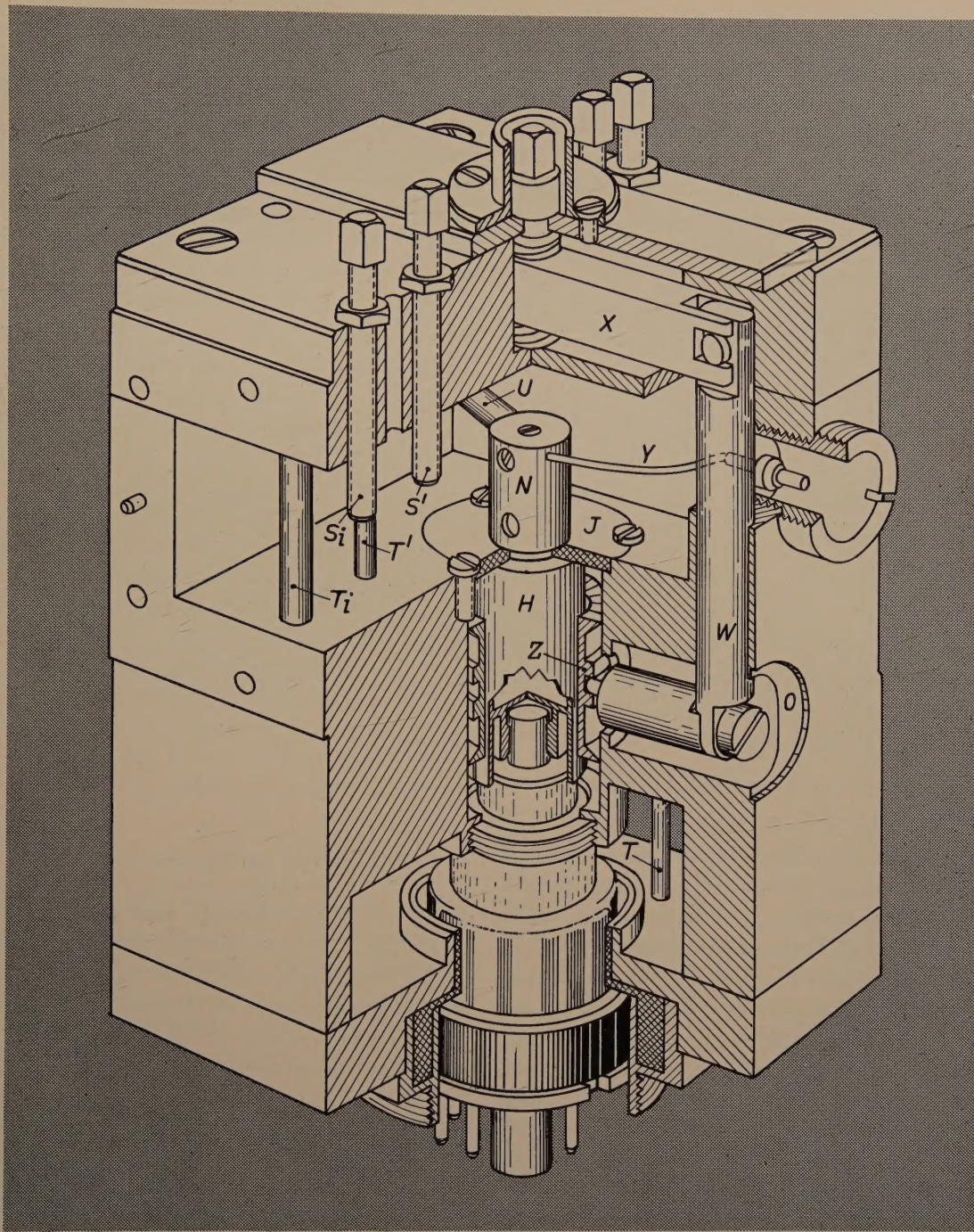
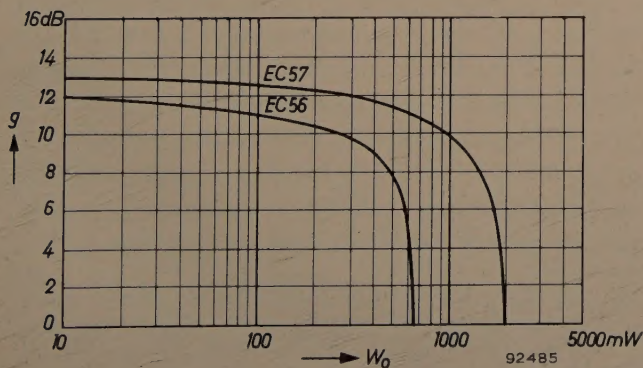


Fig. 13. The complete amplifier block, cut away to show the most important components. W denotes the driving rods for plunger Z . These rods are fixed at pivots to cross-beam X . Underneath they carry pins which engage with the plunger. The other letters have the same meaning as in earlier figures.

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92485

The two causes of feedback oppose each other and at a given frequency, can even cancel each other out. To illustrate this, an equivalent circuit is shown in *fig. 15a* for a tube in which only the heater voltage is switched on. The elements C_{ak} and L_g are responsible for the two feedback effects. Applying a delta-star transformation to the three capa-

Fig. 14. Gain g as a function of the output power W_0 for tubes EC 56 and EC 57, at a bandwidth b_1 of 55 Mc/s.

citors, we obtain the result as shown in fig. 15b. It will be evident that no feedback appears at the resonant frequency of L_g and the equivalent capacitance C_3 in series. With the tubes EC 56 and EC 57 this resonant frequency is at present about 5200 Mc/s and is thus considerably higher than the frequencies at which the amplifier will be used. At a frequency of 4000 Mc/s the series arrangement of L_g and C_3 therefore has a reactance 2.5 times smaller than the reactance of C_3 alone. It may broadly be said, then, that owing to the presence of L_g the capacitance C_{ak} is apparently reduced by the same factor.

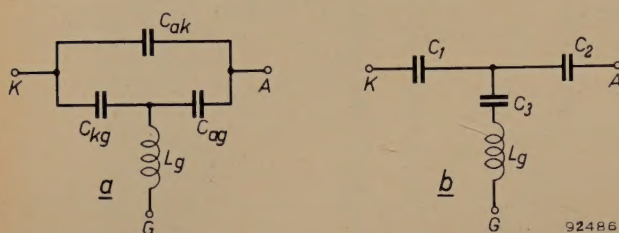


Fig. 15. a) Equivalent circuit for a tube with only its heater voltage applied. A anode, K cathode, G grid, L_g equivalent inductance of the grid wires. b) Diagram obtained by applying a delta-star transformation to the three capacitors.

When the tube is in operation the situation is considerably more complicated, mainly because of the effects of space charge and electron transit time, which increase the input and output damping and also give rise to a phase shift between the voltages on cathode and anode. No complete theory has yet been presented to account for all these facts, but enough is now known to explain some of the phenomena occurring in a tube under operating conditions.

The feedback in the tube need not be regarded entirely as a drawback. As pointed out by Van der Ziel and Knol¹⁰⁾, feedback can be used to increase the product of gain and bandwidth. In fact, the favourable properties of tubes, EC 56 and EC 57 are partly due to feedback.

In another respect, however, feedback does cause difficulties, especially if one wishes to connect several of these amplifiers in cascade. It is then obviously convenient to arrange for the anode circuit of each tube to form a band-pass filter with the cathode circuit of the next tube. When this is done the role of the iris, which we discussed when dealing with the anode circuit, is taken over by the cathode circuit of the next tube. Owing to the feedback, however, the cathode circuit of the tube

can no longer be regarded as a simple resonant circuit. For instance, if the input side of a tube is matched to the input waveguide while the anode resonant circuit is detuned, the tuning of this cavity can seriously upset the matching at the input side. Since the bandwidth of the anode circuit is much smaller than that of the cathode circuit, this interference occurs in a relatively narrow frequency band, so that it becomes very difficult to form with this input circuit a band-pass filter with transitional coupling. The effect is stronger the higher the resonance resistance of the anode resonant cavity and thus the narrower the bandwidth of the anode circuit.

In spite of these difficulties it has proved possible to connect three amplifier stages in cascade and to make the overall frequency characteristic sufficiently flat. One can never be certain, however, that the stages separately have a flat frequency characteristic, and there is accordingly a considerable risk that a change in the characteristics of the tubes will be associated with an unwanted distortion of the frequency characteristic.

The use of several stages in cascade can be made much easier by effecting the coupling between the stages via ferrite directional isolators.

The use of ferrite isolators

For an explanation of how a ferrite isolator functions reference is made to an earlier article in this Review¹¹⁾. The article describes, among other things, an isolator developed for use at frequencies of about 4000 Mc/s. By placing an isolator of this kind, both ends of which are matched to the waveguide, between two amplifier stages, many difficulties arising from feedback are overcome and, moreover, several other advantages obtained. Each stage can now, with its associated isolator, be tuned as a separate unit, and several such stages can readily be connected in cascade. The sacrifice this entails is a slight loss of power, amounting to less than 1 dB per isolator.

Fig. 16 shows three cascaded amplifier stages, coupled via ferrite isolators. Using two EC 56 tubes and one EC 57, the output power of the amplifier is 1.5 W with an input power of 1.5 mW. The total gain is thus 30 dB. With a bandwidth b_t per stage of 55 Mc/s, the bandwidth of the total amplifier is 42 Mc/s. If a tube has to be changed, it is only necessary to readjust the stage in question.

¹⁰⁾ A. van der Ziel and K. S. Knol, On the power gain and the bandwidth of feedback amplifier stages, Philips Res. Rep. 4, 168-178, 1949.

¹¹⁾ H. G. Beljers, The application of ferroxcube in unidirectional waveguides and its bearing on the principle of reciprocity, Philips tech. Rev. 18, 158-166, 1956/57 (No. 6).

When an isolator is employed, the band-pass filter in the anode circuit of the tube is not (as with direct inter-stage coupling) partly formed by the cathode circuit of the following tube, but

frequency; at frequencies differing by 10 Mc/s from the central frequency, the group delay differs by only 64×10^{-12} sec from the above value. In consequence of these favourable figures it may, in

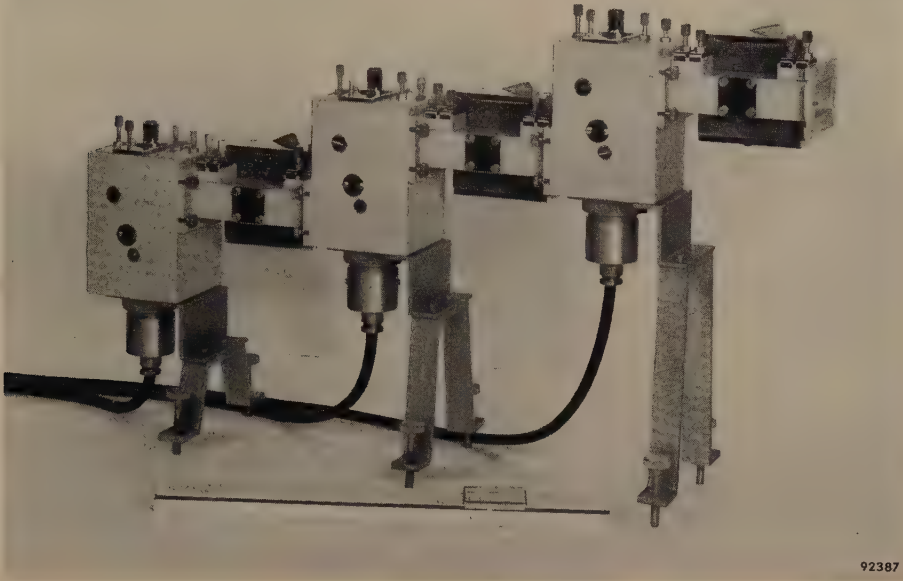


Fig. 16. Amplifier, consisting of three high-frequency stages coupled via ferrite isolators.

by the anode resonant cavity and a waveguide iris, in the manner already described. The input capacitance of the tube then has no effect on the properties of this band-pass filter, and since this capacitance is the quantity that varies most during operation, the shape of the frequency characteristic is better maintained when isolators are used.

Group delay

For the undistorted transmission of frequency-modulated signals it is not only necessary that the amplification is constant over the whole frequency band concerned; it is equally necessary that the group delay in that band should be constant. Measurements and calculations have shown that a high-frequency amplifier, as described here, has much more favourable properties in this respect than a normal intermediate frequency amplifier, as used in most microwave links¹²). This is largely due to the fact that the absolute bandwidth of an IF amplifier is always much smaller than that of a high-frequency amplifier.

The group delay of an amplifier stage such as described above is 3.2×10^{-9} sec at the central

certain circumstances, be advantageous for relay stations in microwave links to use no IF amplifiers but only a number of high-frequency amplifier stages according to the principle discussed. Since in this case the product of gain and bandwidth per stage is greater than in the case of an IF amplifier, it is possible to reduce the total number of stages in the amplifier, thereby considerably reducing the variation of group delay in the transmitted frequency band.

The above-mentioned figures for the group delay refer to a perfectly adjusted amplifier. It should be borne in mind, however, that after some time in operation the amplifier may no longer be exactly adjusted. (One cause may be a change in the inter-electrode capacitances of the tubes.) The usual consequence of this is a change in the shape both of the phase characteristic and the amplitude characteristic. The group delay is, of course, directly affected by deviations in the phase characteristic, but it can also be indirectly affected by changes in the amplitude characteristic, especially if the amplifier delivers a high power output, in which case the tube capacitances depend upon the magnitude of the signal voltages. As a result, an amplitude variation, caused in frequency-modulated signals by the amplitude characteristic having an unwanted

¹²) See J. P. M. Gieles, The measurement of group delay in triode amplifiers at 4000 Mc/s. This article will shortly appear in *L'Onde Electrique*.

shape, can be associated with a phase variation, which in turn gives rise to a change in the group delay. It has been found, however, that the latter effects are very small in the amplifier discussed in this article.

There are certain other less important causes of variations of group-delay in the amplifier, one of them being the electron transit time in the tubes and another the group-delay of the electromagnetic waves in the isolators. We shall not give details here of group-delay measurements carried out on this amplifier, but mention merely that the results of such measurements lead us to expect that it will be possible with 50 relay stations, equipped solely with amplifiers as discussed in this article, to transmit a colour television signal according to the NTSC system over a distance of 2500 km, without exceeding the maximum permissible distortion.

In the development of the amplifier described here, important contributions have been made by G. de Vries and A. Meyer of the Eindhoven Research

Laboratory, H. Kramer of Philips Telecommunication Industry in Hilversum and W. Busscher, J. Jongsma and K. Rodenhuis of the Electronic Tubes Development Laboratory in Eindhoven.

Summary. This article describes the construction and properties of an amplifier for operation at frequencies from 3800 to 4200 Mc/s. The tube used in the amplifier may be an EC 56 or an EC 57, with which an output power of 0.5 and 1.5 W respectively can be obtained with a gain of 8 dB. The low level gain is 12 and 13 dB respectively. The frequency characteristic is flat so that the gain varies by no more than 0.1 dB in a frequency band of 55 Mc/s. The input and output of the amplifier are designed for connection to waveguides. In the anode circuit a band-pass filter is used, which is formed by a resonant cavity coupled with a resonant circuit in the output waveguide. The input and output waveguides contain variable reactive elements for input matching and for adjusting the anode circuit. When several of these amplifiers are connected in cascade it becomes difficult to adjust them correctly, owing to feedback. These difficulties are largely overcome by using ferrite isolators between the stages. The variation of group delay in the frequency band covered is much smaller in this amplifier than in the IF amplifiers normally used in microwave radio links. It may therefore be advantageous for relay stations to use no IF amplifiers but only a number of the high-frequency amplifier stages described here.

A LUMINOUS FRAME AROUND THE TELEVISION SCREEN

621.397.62

People generally prefer to view television in a lighted room. The brightness level in the room, around the set, however, is always considerably below that of the picture. An improvement in viewing conditions appears to be obtained by introducing a zone of intermediate brightness between the screen and its surroundings. Such a transition zone is generally provided by surrounding the picture with a light-coloured frame. The question now arises whether it might not be an improvement to provide this frame with its own illumination. Also, how wide and how bright should such a frame be? An answer to these questions was obtained by means of the following tests.

On a screen of 480×360 mm, which served as a substitute for a TV screen, no TV transmissions being available at the time of the tests, a film was projected by a 16 mm projector. The projector was fitted with a light-blue filter to make the colour of the projected image correspond to that of a TV picture. The projection screen was surrounded with a frame whose luminance and width could be varied with the aid of control knobs by an observer seated at a distance of 2.20 m from the screen (*fig. 1*). The luminance was variable between 0 and 34 cd/m^2 ; the width between 0 and 160 mm. The

surroundings, which were grey or black curtains, were given a known and adjustable luminance, uniform throughout the entire field of vision.

Whilst the observer watched and listened to the film, he was asked — at times when the average screen luminance, measured in advance, remained fairly constant for longish periods — to adjust the frame in accordance with:

- the luminance he preferred at each of four given values of the frame width,
- the width he preferred at each of four given values of frame luminance.

Graphical interpretation of the results made it possible to establish which combination of frame luminance and frame width the observer preferred for the various given values of the average screen luminance at three different luminance levels of the surroundings (see *Table*). Care was taken that

Table. Combinations of luminance values of the surroundings and average luminance values of the screen (in cd/m^2) at which the adjustments of frame width and frame luminance were carried out.

Surroundings	0.005			0.5			5.0	
Screen	2.0	7.3	30	3.8	9.1	32	25	48

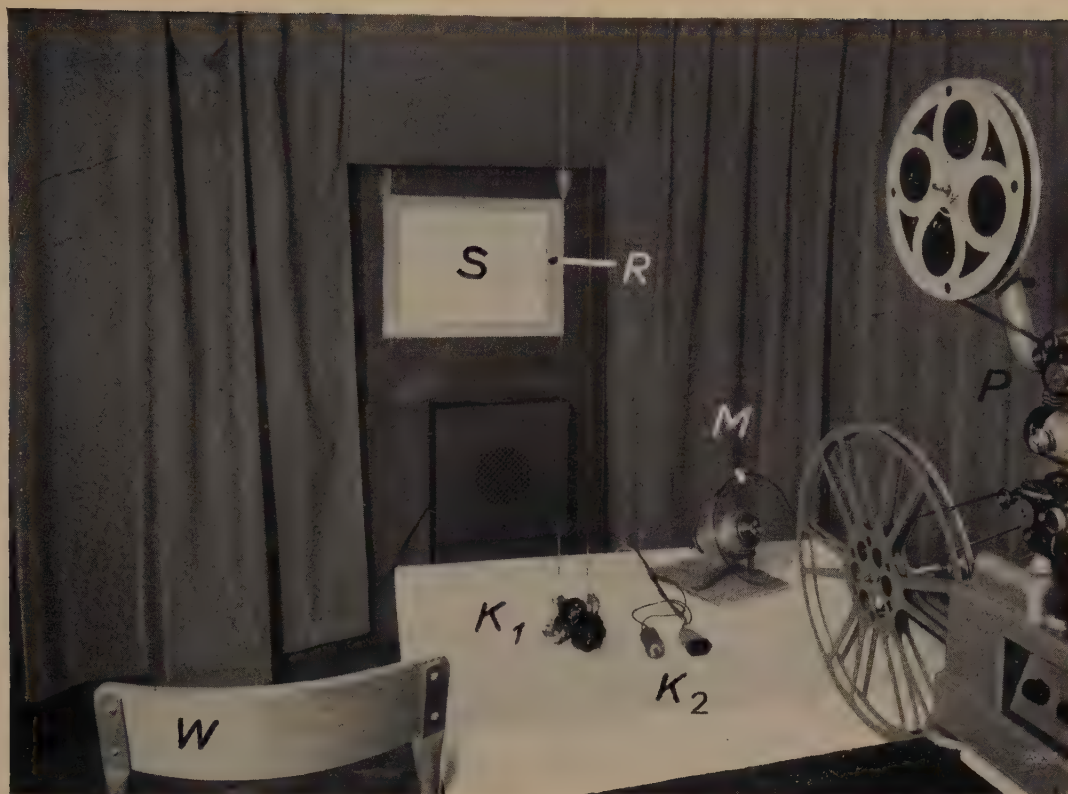


Fig. 1. Test arrangement for determining the most favourable width and brightness of the frame around a TV screen. A substitute for a TV picture is produced on the screen *S* by the film projector *P*. The observer, seated on chair *W*, can operate knob *K*₁ to adjust the luminance of the frame *R* around the screen, and the knobs *K*₂ (one being used for reversing the movement) for varying the width of this frame by means of two sliding masks. The ratio of the width of the frame along the short edge of the screen to that along the long edge remains constant at 4 : 3. The adjusted luminance can be read from disc *M* by the person conducting the test, who also measures the adjusted frame width. The desired luminance of the surroundings is obtained by a variable arrangement of uniform diffuse lighting and by variation of the reflection factor of the surroundings (other curtains).

no one observer saw the same film twice, in order to preclude above-normal attention to the frame due to lack of interest in the picture. The observer was explicitly told to carry out the adjustments whilst looking at the picture. For each set of luminance values of screen and surroundings (see Table), the adjustments mentioned under (a) and (b) were carried out by 20 to 25 observers. This provided us with the same number of diagrams of the kind shown in *fig. 2*, and the same number of points *Q* representing preferred combinations of frame width and frame luminance. *Fig. 3* shows the cluster of points obtained for a screen luminance of 25 cd/m² and a surroundings luminance of 5 cd/m². There is evidently a considerable difference between the individual preferences of the various observers.

The following conclusions can be drawn from the tests¹⁾. As regards the preferred frame width, no correlation was found with the luminance of the surroundings of the screen. The average preferred

frame width over all the tests was 0.3 times the half screen dimension. This value lies close to the customary widths of frames on modern TV receivers.

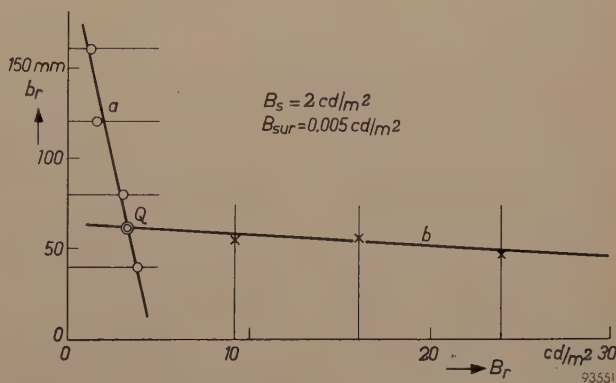


Fig. 2. For each of the four given frame widths *b*_r, the observer adjusted the frame luminance *B*_r to the level he preferred (circles), and for each of the four given frame luminosities the same observer adjusted the width of the frame to the value he preferred (crosses). The intersection *Q* of line *a* (through the circles) and line *b* (through the crosses) represents the combination of frame luminance and frame width most preferred by this particular observer for the prevailing average luminance *B*_s of the screen and the luminance of the surroundings *B*_{sur}.

¹⁾ A more detailed account of these tests and of the statistical evaluation of the results is published in *Light and lighting* 50, 245-250, 1957 (No. 8).

The preferred frame luminance was found to increase with the screen luminance as well as with the luminance of the surroundings (fig. 4). For modern TV sets in lighted rooms, it appears that a frame luminance somewhere between 10 and 20 cd/m² would be preferred. A statistical analysis of the test results further demonstrates that the diverging wishes of the large majority of viewers can be fulfilled by making the frame luminance variable between 0 and 40 cd/m².

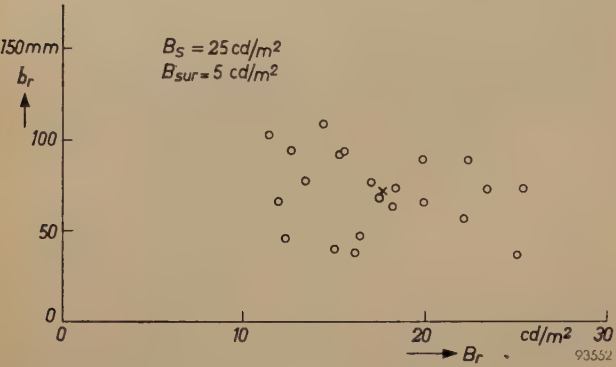


Fig. 3. Example of a cluster of points consisting of all points Q derived in the manner shown in fig. 2 (each observer supplying one point). This scatter diagram, which clearly illustrates individual differences, is, like fig. 2, only valid for a given set of luminance values of screen and surroundings. The cross shows the average preferred combination.

The customary non-illuminated screen frames derive their brightness from light reflected from the room and from the screen. Light-coloured frames might sometimes attain a luminance of 10 cd/m²;

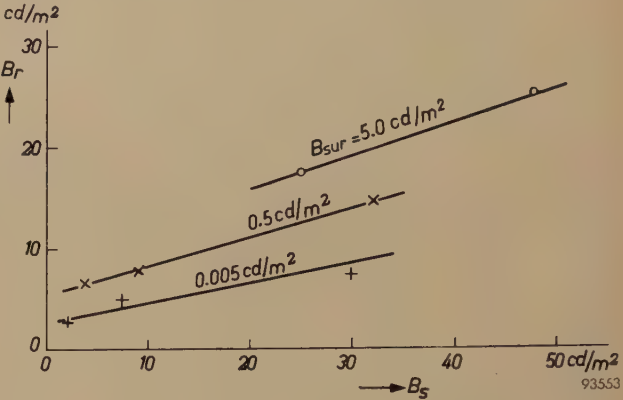


Fig. 4. Average values of the preferred frame luminances B_r as a function of the average screen luminance B_s , for three values of the luminance B_{sur} of the surroundings.

for a dull, fairly dark gray frame, 5 cd/m² is about the upper limit. We may conclude from this that separate illumination of the frame might sometimes be worth while.

As regards colour, the observers unanimously preferred a frame of the same colour as the picture.

J. J. BALDER.

LOW-VOLTAGE OSCILLOSCOPE TUBES

by F. de BOER and W. F. NIENHUIS.

621.317.755

The demand for cathode-ray oscilloscopes has greatly increased in the last decade. In addition to the demand for comprehensive instruments the need has arisen for small sets such as are employed in radio and television servicing workshops. It would be possible to effect a considerable reduction in the weight and bulk of oscilloscopes if only the components generating the voltage required for the final anode of the CRT could be made smaller. In this article a type of CRT is described which requires an anode voltage very much lower than usual, with a consequent reduction in the size and weight of the oscilloscope. Despite the low voltage on its anode the tube gives a good clear image and offers many possibilities; it will appeal to radio amateurs, amongst others.

Most oscilloscope tubes in common use require from 800 to 1500 volts on their anodes: one consequence of this was that all oscilloscopes have had to embody a heavy, bulky and expensive high tension transformer. An obvious first step towards building the desired small lightweight oscilloscopes was to try to design a CRT that would work satisfactorily with a low anode voltage. Lowering the anode voltage has the additional advantage that the sensitivity of the tube is increased, thus reducing the amplification that the test signal has to undergo; the appropriate circuits can then be simplified.

We have now succeeded in making such tubes. The DG 7-32 and DG 7-31, which will be described below, require a voltage of only 400 V (minimum).

The use of an anode voltage as low as this means that the composition of the fluorescent layer and the manner in which it is applied have to meet highly specialized requirements; the electrode assembly too has to be suited to the low anode voltage if good resolution (i.e. a small enough spot) is to be obtained.

The nature of these problems and the solutions found for them will be discussed below. A small cathode-ray oscillograph making use of a tube of this type is described briefly at the end of the article. It should be noted at this juncture that the DG 7-32 tube has been designed for a sweep voltage that is symmetrical with respect to earth; in the DG 7-31 tube one of the plates is earthed, and the sweep voltage is therefore applied asymmetrically. The two tubes are of the same size and have a screen diameter of 7 cm.

The fluorescent screen

For a tube working at these low anode voltages, the fluorescent screen cannot be fixed to the glass of the envelope with the aid of some soluble silicate

binder¹⁾, as is the normal practice. Although the thickness of the film of binder lying over the screen is only of the order of 0.01 micron, electrons of about 400 eV lose more than 95% of their kinetic energy in passing through, with the result that the brightness of the trace is cut down excessively. Again, any chemical action on the grains of the fluorescent powder, such as might take place during the application of the screen, would be disastrous for the same reason.

This being so, the old and familiar "flow coating" method is the one preferred. A certain quantity of a suspension of the fluorescent powder in a solution of nitrocellulose in some organic solvent (e.g. ethyl acetate) is poured into the envelope. By rotating the tube the entire screen surface is covered with the liquid and then allowed to dry off. Subsequently the nitrocellulose is removed by heating up to 400 °C. Owing to the volatility of the decomposition products and the moderate temperature the fluorescent powder undergoes no chemical change.

It is well known that a layer of this kind cannot be used in conjunction with a low anode voltage without provision being made for the fact that the electron beam is not adequately balanced by secondary emission from the screen, which consequently acquires a charge²⁾. Moreover, covering the fluorescent layer with either a conductive layer or a layer having a suitable coefficient of secondary emission (as is done in television picture tubes) is out of the question for the reasons already stated. However, the screen can be prevented from charging up by previously giving the screen portion of the envelope interior a transparent conductive

¹⁾ F. de Boer and H. Emmens, Philips tech. Rev. **16**, 232-236, 1954/55.

²⁾ Cf. J. de Gier, A. C. Kleisma and J. Peper, Philips tech. Rev. **16**, 26-32, 1954/55.

coating that serves as base for the fluorescent powder. Tin oxide with certain additives is a suitable substance for the coating, conferring on it the properties of a semiconductor³⁾. The coating is applied by spraying a tin chloride solution into the tube, which is heated to temperature approaching the softening point. The conductive coating only "takes" where the temperature of the glass is sufficiently high; hence the operative has the place where it forms well under control.

An incidental but important advantage of having a conductive base for the screen is to prevent the screen potential being affected should the tube be touched when, for example, measurements are being made. There is therefore no need for the screen to be at earth potential, as was usually the case in earlier types of oscilloscope tube. Indeed, in the new types the cathode of the electron gun can be earthed, so that the heater can be fed from one of the normal heater windings provided on the mains transformer.

Coatings of this kind generally give rise to interference colours. Their formation might be prevented by making the depth of the coating less than a quarter-wavelength of violet light, but so thin a coating would have insufficient conductivity for our purposes. Nor, on the other hand, can one make the coating so thick that interference would cease to play any part, because then it would not adhere to the glass. The coating that is in fact applied has a thickness such that no interference colours of an order higher than the first are produced. Furthermore, in order to avoid colour differences, this depth is kept as nearly constant as possible throughout the screen area. It is only the outward appearance of the tube that is concerned here; the interference effect makes no perceptible difference to the oscillogram itself.

The fluorescent powder must also be selected to suit the low anode voltage. From the brightness viewpoint the best phosphor is normal willemite (zinc orthosilicate activated with manganese in a Si/Mn ratio of 1 : 0.008). However, when a powder of normal composition is used, it is found that after about 600 hours the fall-off in brightness due to electron bombardment is steeper per unit of time than is considered desirable — see graph *a* in *fig. 1*. When an oscillogram on a tube of that age is examined, the points at which it intersects traces of earlier oscillograms are perceptible as points of lower brightness. The steep decline in brightness can be prevented by raising the manganese content; but then the initial brightness falls off. The relationship between the two quantities is shown graphically in *fig. 2*. The best compromise was found to be a

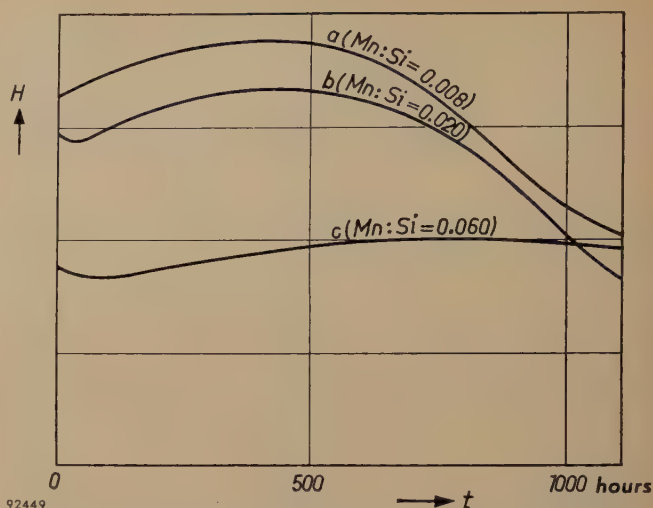


Fig. 1. Curves of H , the brightness of a trace on a willemite screen, as a function of time, when the screen is bombarded with electrons of 400 eV. The ratio of manganese to silicon atoms in the phosphor is 0.008 : 1 (the normal ratio) for curve *a*, 0.020 : 1 for curve *b* and 0.060 : 1 for curve *c*.

Mn/Si ratio of between 3 : 100 and 4 : 100. This gives curves lying in the region between curves *b* and *c* in *fig. 1*. The resulting degree of brightness is not unduly low (as it is in the case of curve *c*), nor does it fall off too quickly with time (as it does in curves *a* and *b*).

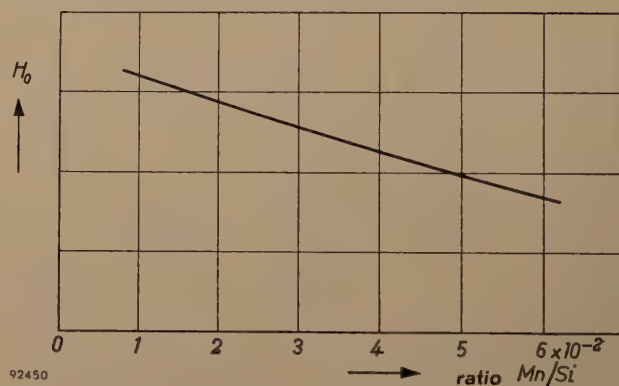


Fig. 2. Initial brightness H_0 of willemite screens as a function of the manganese content of the phosphor. The vertical axis is divided into arbitrary brightness units and the horizontal axis according to the ratio between manganese and silicon atoms. The curve shown is that obtained with a beam of 400 eV electrons.

The electrode assembly

A diagram showing the electrode assembly of the DG 7-32 appears in *fig. 3*. The assembly has three main parts, an electron gun delivering a beam of adjustable intensity, an electron lens that brings the image of the beam cross-over⁴⁾ to a sharp focus on the screen, and a deflecting system whereby the beam is made to undergo the desired changes of direction.

⁴⁾ The cross-over is the point at which the diameter of the beam in the gun is smallest; it lies somewhere between the cathode and first anode. Refer to *fig. 5* of article cited in footnote ⁵⁾.

³⁾ See for example, H. te Gude and E. Schaaff, Philips tech. Rev. 18, 243-245, 1956/57 (No. 8).

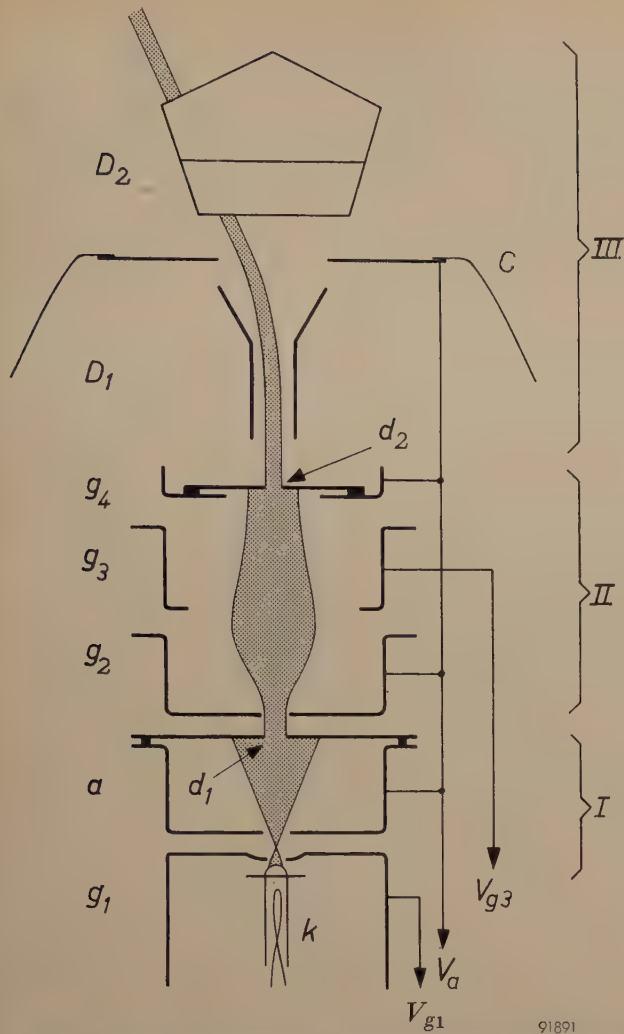


Fig. 3. Diagram to show the electrode assembly of a DG 7-32 oscilloscope tube. Three consecutive sections of the assembly may be distinguished, (I) the electron gun, (II) the electron lens, and (III) the deflecting system. Diaphragms d_1 and d_2 are electrically connected to the electrodes on which they are mounted. It will be observed that electrodes a , g_2 and g_3 have the same basic shape, a certain degree of standardization having been achieved in the parts making up the assembly.

A photograph of the tube, together with a complete and finished electrode assembly and the individual parts of which it is made up, appears in fig. 4.

Electron gun and lens

A triode gun is used, consisting of an indirectly heated cathode k , a control grid g_1 and a first anode a . The cathode is earthed, g_1 carries negative potential and the anode a has a potential of $+400$ V. The intensity of the beam can be regulated⁵⁾ by changing the potential of g_1 .

Electrodes g_2 , g_3 and g_4 form the electron lens. By changing the potential of g_3 (the focussing electrode) the strength of the lens can be adjusted so as to give the sharpest possible image of the crossover on the screen, that is, so as to produce the

⁵⁾ Electron guns of all types are dealt with in J. C. Francken, J. de Gier and W. F. Nienhuis, Philips tech. Rev. 18, 73-81, 1956/57 (No. 3).



Fig. 4. A DG 7-32 tube, a finished electrode assembly and the individual parts comprising the assembly. The tangs on the grids and the D_1 plates can clearly be seen; these tangs are lodged in the powdered-glass filling of grooves in the ceramic rods.

smallest possible spot. The electrodes g_2 and g_4 have the same potential as a . The electrode g_3 carries a potential between 0 and +200 V, depending on the anode voltage.

It will be seen that in these tubes the functions of shaping and focussing the beam are distributed amongst several electrodes; this system compared with others that were formerly much used (systems with accelerating lenses), has the great advantage that the intensity and sharpness of the spot can be adjusted independently of each other.

Diaphragms d_1 and d_2 have the following function. d_1 constricts the beam issuing from the gun, thus preventing electrons striking the focussing grid g_3 . Thus focussing is effected without a flow of current, making it permissible for the voltage source supplying g_3 to have a high internal resistance. Electrons which are scattered around the edges of d_1 and which might otherwise strike g_3 are intercepted by g_2 . Diaphragm d_2 constricts the beam prior to its entry into the deflecting system.

Having been narrowed down thus by the agency of d_1 and d_2 , the beam has only a small fraction of its original intensity on arrival at the screen. The loss of beam strength is particularly undesirable in a CRT with a low anode voltage, where the production of a sufficiently bright spot is a problem in itself. In order to reduce the loss the beam must be made as narrow as possible before it reaches d_1 , and this may be done by strengthening the lens formed by g_1 and a . Doing so, however, is only too likely to lead to enlargement of the spot on the screen⁶⁾. Enlargement of the spot may be prevented by bringing the lens nearer to the screen, but then the deflecting system has to be moved up too and this in its turn leads to a loss of deflection sensitivity. A compromise has therefore to be sought. In the tubes under discussion, the g_1 - g_2 lens is given somewhat greater strength than the corresponding lens of the guns described in article⁵⁾ by making a depression in the face of g_1 .

Deflection system

The deflection system does not differ in principle from that which has been employed in oscilloscope tubes for some time now. We shall briefly discuss two details which have not received particular attention in previous articles. The two pairs of plates, pair D_1 for vertical deflection by the test voltage and pair D_2 for horizontal deflection, may be seen in fig. 3. In a tube with flat parallel deflecting plates (as in fig. 5) the formula for R , the displacement of the spot across the screen, is:

$$R = \frac{Ll}{2V_0S} V_d, \dots \dots \dots (1)$$

where V_0 is the difference of potential through which the electrons have fallen, V_d is the difference of potential between the plates, l the length of the plates, S their distance apart and L the distance from their centre to the screen.

It follows directly from the formula that, where V_0 and L and the screen diameter are given, the plate voltage required for maximum deflection can be lowered by increasing the ratio l/S . This can only be done to a limited extent, however, because a) the beam diameter d (see fig. 5) cannot be narrowed too much without reducing the brightness

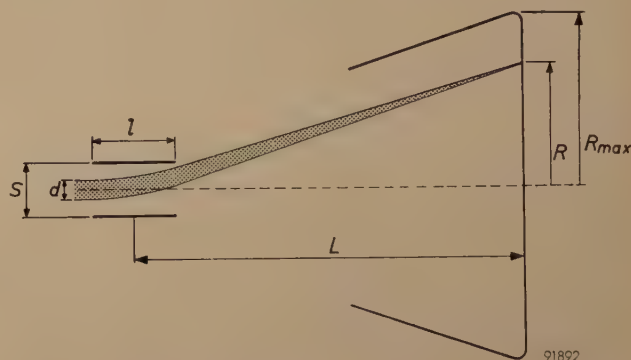


Fig. 5. The deflection of an electron beam by flat parallel plates.

of the spot, and b) the beam will soon start to be pulled against the plates if l is lengthened unduly, and this will result in distortion of the trace. The aim of lowering the deflecting voltage can nevertheless be achieved by bending the plates outwards; the bend in the D_1 plates can be seen in fig. 3, that in the D_2 plates in fig. 4.

An improvement of more recent date is a change in the shape of the D_2 plates; these are not rectangular, as before, but have the outline indicated in fig. 3 (see also fig. 4). The aim achieved by giving the D_2 plates this shape is that the path of the electrons between them has the same effective length whatever the degree of deflection they have previously undergone between the D_1 plates; in other words, the amount of deflection undergone in D_2 is independent of that undergone in D_1 . Pincushion distortion of the image is thereby obviated.

One inference from formula (1) that is of particular importance for tubes with a low V_0 is that R/V_d , the sensitivity of the deflecting system, is inversely proportional to V_0 ⁷⁾. For previous types of CRT the minimum value of V_0 has been 800 V; the reduction of V_0 to 400 V, as in the present tubes,

⁶⁾ A fuller account of "pre-focussing" may be found in the article cited in footnote⁵⁾.

⁷⁾ It is true that the formula has to be somewhat modified in respect of the bending of the plates, but this does not affect the part played by V_0 .

therefore results in doubled deflection sensitivity. The average sensitivity of these tubes, both the DG 7-31 and the DG 7-32, (with V_0 at 400 V) is approx. 0.50 mm/V for the D_1 plates, which carry the test voltage, and approx. 0.30 mm/V for the D_2 plates.

Mounting of the electrodes

The electrodes are mounted on ceramic rods having a groove filled with powdered glass, in a manner that has already been described in this Review⁹). The rods are heated to a temperature



Fig. 6. An oscilloscope of small size, equipped with the DG 7-32 tube (this instrument is not on the market). The properties referred to in the text have been obtained for the oscilloscope in the following way. The incoming signal is passed through a two-stage amplifier equipped with two ECF 80 triode-pentodes. The triode portion of the first tube, connected as a cathode-follower, precedes the pentode portion; the second triode serves as a phase inverter. Even better properties can be conferred on the oscilloscope by using an attenuating probe in conjunction with it. In the first place, of course, the probe allows a stronger test signal to be admitted, but it also makes it possible to reduce the input capacitance to 5 pF. For this purpose the ECF 80 is replaced by a combination of an EF 80 pentode and an EC 92 triode. The latter tube, again as a cathode-follower, is embodied in the probe. With the CRT operating with the minimum sensitivity of 0.27 mm/V (i.e. with 400 V on the final anode), the amplitude of the sweep voltage required for a time axis 70 mm long is 126 V. The sweep voltage is generated by an EF 80 tube in a Miller transitron circuit¹⁰), one half of the ECC 81 double triode acting as phase inverter. The other triode acts as limiter for the synchronizing signal.

In the DG 7-31 tube, which has been designed for asymmetrical application of the sweep voltage, the unearthed plate has two metal rods fixed to it on the side nearest the D_1 plates. The function of these rods is to correct trapezium distortion due to the fact that the mean potential difference between the two pairs of plates D_1 and D_2 is not equal to zero⁸).

Asymmetrical application of the sweep voltage enables the circuit supplying that voltage to be simplified. It does, however, have the drawback that some slight distortion is unavoidable.

⁸) See J. D. Veegens, Philips tech. Rev. 4, 198-204, 1939.

such that the glass melts, permitting the tangs of the electrodes to be pressed into the grooves.

Besides acting as a shield, plate C (see fig. 3) serves for centering the electrode assembly and is provided with springs for that purpose. In the

⁹) J. de Gier and A. P. van Rooy, Philips tech. Rev. 9, 180-184, 1947/48.

¹⁰) See for example J. Czech, The cathode-ray oscilloscope, Philips Technical Library 1957, page 77.



92388

Fig. 7. An oscillogram displayed on one of the tubes described in the article (actual size).

DG 7-31 tube the compensating rods fixed to D_2 prevent plate C being mounted in the place indicated in fig. 3. In this tube, therefore, the centering plate is combined with grid g_4 . Apart from this, the

electrode assemblies of the two tubes are the same.

As already stated, the tubes just described can be made the basis of a very compact and lightweight oscilloscope such as may serve the purposes of television servicing, for example. A photograph of a small instrument of this kind, equipped with the DG 7-32 tube, appears in *fig. 6*. Its dimensions are only $10 \times 12 \times 27$ cm, it weighs 2.6 kg and its power consumption is as low as 26 watts ¹¹⁾.

With a test voltage amplifier having a bandwidth ¹²⁾ of 3 Mc/s, the oscilloscope has a sensitivity of 100 mV per cm. The sweep frequency can be varied from 20 c/s to 25 kc/s. The maximum amplitude permissible in the input signal is 15 V. Input capacitance is 20 pF.

An oscillogram has been reproduced in *fig. 7* to give an impression of the performance of these tubes. In particular, the extremely good definition of the trace is noteworthy.

¹¹⁾ The instrument was developed by J. Stolk and W. D. Minjon in the Eindhoven applications laboratory for cathode-ray tubes.

¹²⁾ Defined, as is usual, as the width of the band between those frequencies at which the amplitude falls off by 3 dB.

Summary. An account is given of the development of oscilloscope tubes requiring an anode voltage of only 400 V. There are two types, DG 7-32 and DG 7-31. The sweep voltage is fed to the first type symmetrically; one X-plate is earthed in the second type, the sweep voltage therefore being fed asymmetrically. The fluorescent screen is applied by the flow-coating method, and not with the aid of an inorganic binder. Such a layer of binder, covering the screen, would decelerate the beam electrons excessively. To prevent the screen from becoming charged, the screen portion of the envelope is first given a conductive coating of tin oxide. The coating offers two additional advantages: in the first place, when measurements are being made, the glass can be touched without the screen potential being affected; secondly, it is possible for the

cathode of the tube to be earthed, so that its heater can be fed from one of the normal heater windings on the mains transformer. The electrode assembly has been designed in such a way that focussing of the beam and adjustment of its intensity can be carried out independently of each other. The spot produced on the screen is small, and the definition of the trace therefore good. Two improvements to the deflecting system, introduced some time ago, are dealt with. Finally, some details are given of an oscilloscope of very light weight and of very small dimensions, the construction of which has been rendered possible by the availability of the tubes here described. The possibility of making such an instrument will be of particular interest to the radio and television serviceman, and also to the amateur equipment builder.

THE FORMULATION OF PESTICIDES

by W. DUYFJES *).

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Agriculture and horticulture are increasingly exposed to plagues of such organisms as insects, mites, fungi and weeds. The chemical means of combating such pests come under the general heading of "pesticides". In previous volumes of this journal, articles have described the research work conducted by N.V. Philips-Roxane in the quest for new pesticides. The value of a pesticide depends not only on the active substance; the "formulation" of the preparation is equally important. By formulation is understood the making up of the active substance in a form (solution, emulsion, suspension, etc.) in which it is most efficacious, stable in transport and storage, reasonable in price, and so on. The problems relating to formulation are the subject of the present article and are studied in the formulation laboratory at the Amsterdam factory of N.V. Philips-Roxane.

The chemical control of agricultural pests

The great increase in world population in recent decades is largely to be attributed to the rapid advance of science in the widest sense of the word. Chemistry has had a large share in that advance: new medicaments have saved countless human lives, and the use of artificial fertilizers has made it possible to put vast barren regions under cultivation. This intensification of agriculture and horticulture has led to considerably increased food production, but unfortunately it has also had an undesirable consequence: injurious organisms, such as certain insects, red spiders, moulds and weeds have found new breeding grounds and threaten to rob man of what he has newly acquired. Although small mammals, birds, amphibians, reptiles and spiders take enormous toll of injurious insects, this is not nearly enough to avert the danger. Mechanical methods of extermination were soon found to be no longer an economic proposition, and thus man has fallen back on chemical agents: pesticides. This collective name covers insecticides, acaricides, fungicides and herbicides, specific as their names imply, for a certain type of pest. Chemicals were used in the last century on a very modest scale for destroying insects, moulds and weeds. But it was the discovery by Müller in 1939 of the insecticidal activity of DDT (dichloro-diphenyl-trichloroethane) that gave fillip to the large-scale comparative investigations into the biological activity of known compounds. New insecticides discovered in this search have largely superseded the "natural"

insecticides (nicotine, rotenone, pyrethrins) as well as the synthetic products, such as inorganic arsenates and fluorides used prior to 1939, and have given birth to a new chemical industry. The latter supplies thousands of tons of pesticides a year which contribute towards safeguarding the harvest.

As regards the control of insects, it was found that the presence of chlorine in an organic molecule could lead in certain cases to insecticidal activity. As a corollary to this observation, such substances as BHC (benzene hexachloride), heptachlor, toxaphene, aldrin, dieldrin, endrin, etc. came into use, following upon the discovery of DDT.

A new group of insecticides that was discovered comprised esters of phosphoric and thiophosphoric acids. These form the basis of such substances as tetraethyl pyrophosphate, parathion, malathion, etc., which have rapidly gained ground. The surprising activity, as it was initially described, of DDT and other chlorine-containing insecticides is even surpassed by that of the phosphates and thiophosphates, since they have a wider action spectrum, being also effective against red spider. This was discovered by Schrader just in time, for the red spider had multiplied enormously owing to the use of chlorinated hydrocarbons (DDT, BHC, lindane, etc.) which had decimated its natural enemies.

Such upsets of biological equilibrium resulting from extermination of the enemies of noxious animals, together with the resistance phenomena that have been observed, have led to the realization that the frequently rigorous methods used for combating insects make an early solution to the insect problem unlikely.

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If, initially, the search was for substances that would kill as many types of injurious organisms as possible, the experience gained soon showed that substances with a more selective action, and above all a more intelligent employment of agro-biologicals, made it more probable that the desired goal would be reached. Thus, for example, emphytic (sometimes called systemic) agents have been developed, such as "Systox" and "Pestox", which are assimilated by the plant, rendering it toxic to the parasite. Further, specific ovicides have been discovered: e.g. "Tedion V18" (2,4,5,4'-tetrachlorodiphenyl sulphone)¹⁾ for exterminating red spider eggs; and also insecticides and acaricides with other active groups in the molecule. These new substances were developed not only with a view to getting round certain disadvantages of the known substances, e.g. their tendency to arouse resistance, their toxicity to man and domestic animals, but mainly with the intention of sparing useful organisms (birds, spiders and other natural enemies of the pest), so as to disturb the complex balance of nature as little as possible.

Good results have also been recorded in the combating of fungi and moulds²⁾. Fungicides, such as sulphur, sulphur compounds and copper compounds which before 1939 were the only ones in use, have been supplemented by another important group comprising the dithiocarbaminates, organic mercury compounds, trichloromethyl derivatives and triazines. Fungicides such as the proprietary products zineb and captan have made general headway. Thus far there have been no indications that the new fungicides will create problems like those arising with the insecticides.

The same applies to weed control. Rapid progress in this field is the outcome of the discovery of the "growth-substance herbicides"³⁾, which are marketed under names such as MCPA, 2,4-D and 2,4,5-T.

The importance of the right "formulation"

In practice a relatively small amount of the active substance, the pesticide, must be distributed over the plants that are to be protected against animal pests or moulds, or over the weeds that are to be destroyed. The pesticide can be distributed over a large area by sprinkling or spraying (a liquid),

by dusting (a dry powder) or by atomizing (a liquid). Accordingly, we thus speak of sprays, dusts and aerosols.

In the preparation of a product for one of these modes of application, the pesticide must be mixed with various accessory substances to give a proprietary preparation which is either ready for use or can be made up for use in a simple manner (e.g. by adding water). The main accessory substance is the carrier; others, as we shall see, are wetting agents, dispersing agents, and stickers.

The selection of these substances and the compounding of them, i.e. the formulation of the active ingredient with certain other substances to give a preparation combining the best properties, is of the utmost importance. As a first requirement, an agro-biological preparation must be capable of being distributed over a large area in one of the above-mentioned ways. Moreover it must not corrode, block or cause wear to the nozzle of the equipment. Once on the plant, the preparation must adhere long enough to do its work; it must not be too volatile therefore; nor must it drain off too quickly or be removed by the wind and rain. Naturally the preparation must not deteriorate in storage and should be relatively inexpensive.

Very briefly we shall now describe the train of events in pesticide research at Philips-Roxane⁴⁾. In the Weesp (Holland) laboratory many substances are synthesized which might conceivably possess a pesticidal action. These substances are studied in the "Boekesteyn" agro-biological laboratory for their pesticidal action and those that show promise as insecticides or fungicides are tested for their phytotoxicity. Their toxicity to warm-blooded animals is likewise ascertained. If the results of these tests are favourable (and this is only very seldom the case!) further biological tests and preliminary field assays are undertaken with the substance compounded in a provisional formulation. If the results are satisfactory here also, and it is decided to manufacture the compounds, the question of the proper formulation arises. The form of the preparation and the choice of accessory components can only be arrived at when various properties of the active ingredient are known: vapour pressure, melting point, solubility, milling properties, sensitivity to light, tendency to reduction or oxidation, crystallization properties, hydrolyzability, etc.

Incorrect formulation will make a useless product of a good pesticide. The composition of a prepara-

¹⁾ J. Meltzer, Research on the control of animal pests, Philips tech. Rev. 17, 146-152, 1955/56.

²⁾ M. J. Koopmans, Fungicide research, Philips tech. Rev. 17, 222-229, 1955/56.

³⁾ R. van der Veen, Growth substances in plants, Philips tech. Rev. 17, 294-298, 1955/56.

⁴⁾ R. van der Veen, "Boekesteyn", the agro-biological laboratory of N.V. Philips-Roxane, Philips tech. Rev. 16, 353-359, 1954/55, in particular pp. 357-358.

tion therefore calls for special attention. A survey of the principal problems that arise in formulation now follows.

General considerations on formulation

Particle size

A pesticide can only answer its purpose if the organism it is to control comes into contact with the active substance and takes up sufficient of it. Both conditions are best met if the substance is finely divided. The smaller the particles, the greater their active surface. For example, if grains with an initial diameter of 1 mm are ground down to particles having a volume 10^6 times smaller, i.e. a diameter of $10\ \mu$, these new particles will have a collective surface 100 times greater than the collective surface of the original grains. The probability that the compound will come into contact with the organism is increased to the same extent. Moreover the organism can more readily take up the smaller particles.

How small the particles must be in order to be sufficiently active depends upon all kinds of factors: the active substance, the organism it is to control, the manner in which it is to be applied and the climatic conditions. In general, however, it can be said that preparations in which the particles of active substance are greater than $50\ \mu$, will not be very effective. This does not of course apply where the preparation is to be used in the form of a (true) solution. 10 to $30\ \mu$ is usually a satisfactory diameter for insecticide particles: because of their usually high mobility, insects take up a relatively large amount of a given substance. For the control of mites and particularly their larvae or eggs a finer division must be aimed at, since these organisms are far smaller than most insects and less mobile.

The combating of fungi calls for still greater dispersion. A particle size of only a few μ is the norm for fungicides that are to be applied in suspension or as a dust.

A very fine state of division of the substance has disadvantages, however. The increase in surface that attends finer division makes for more rapid evaporation. This is especially important in the case of insecticides having a residual activity and which must leave a residue on the plant. Some of these have an appreciable vapour pressure, thus too fine a division of the preparation would mean that the protective action was of too short duration. (It should be remembered in this connection that milling never gives a uniform product: if a product

is ground to a mean particle size of say $10\ \mu$, a large number of much smaller particles will be formed which evaporate relatively quickly.)

Volatility can, moreover, bring numerous undesirable reactions to light. If there is any "chemical incompatibility" between the accessory components (more about this later) or some degree of toxicity to the plant which is to be protected, these effects will be magnified with smaller particles. This is true also of the lumping of powder preparations when stored for lengthy periods, especially under heightened pressure (stacked up in bags) and tropical temperatures. The lumping can lead to blockage of the nozzle; another ingredient is therefore necessary to prevent lumping. The World Health Organization has laid down standards to which certain agro-biological preparations have to conform. They include a "tropical storage test"; the success with which a preparation survives this test gives an idea of its ability to stand up to transport and storage in the tropics.

The carrier

The carrier has already been mentioned as the most important accessory component. Its function in many cases is to afford better distribution of the active ingredient. Frequently it also serves as the grinding medium and as an adsorbant which gradually dispenses the active ingredient (this process must be neither too quick nor too slow). Laboratory tests can give valuable indications as regards the rate of dispensation, but in the case of powder preparations it is always necessary to make biological tests to ascertain whether the activity is near enough to the optimum.

In the development of a preparation, the "chemical incompatibility" alluded to above, must be taken into account. An insecticide such as DDT can react with OH-ions, splitting off HCl to give a compound which is biologically inactive. Thus such an insecticide must not in general be formulated with an alkaline carrier (in certain cases however basic carriers are nevertheless compatible with pesticides that split off HCl). Conversely a pesticide may undergo chemical decomposition owing to certain impurities in the carrier which initiate catalytic decomposition. For the above reasons, a compatibility test — which is simple and rapid to carry out — should never be omitted.

Some carriers contain abrasive ingredients. If they possess a certain structure and grain size, they can do such damage to the insect cuticula that water is lost by the insect through evaporation,

with resultant death of the insect. Such carriers therefore possess an insecticidal action in themselves, but are nevertheless highly undesirable, killing useful insects and causing mechanical damage to the plant, and rapid wear of the dusting equipment and spray nozzles.

Powder preparations can be divided into dusts and wettable powders. These differ widely in their contents of active ingredient: dusts are preparations ready for use and thus possess a fairly low content of active substance; on the other hand wettable powders are suspended in water before use and the actual ("secondary") carrier is thus water. For reasons of economy (freight costs), the content of active substance in wettable powders is made as high as possible. The ("primary") carrier present in the powder must thus possess a much greater adsorbing capacity, and consequently the danger of lumping (see above) is appreciably more imminent. The formulation of wettable powders therefore demands greater care than the formulation of dusts.

Other ingredients

On account of the water-repellent nature of many pesticides their suspension in water calls for a wetting agent, i.e. a substance that lowers the surface tension of water. This wetting agent must also be compatible with the active substance and with both the primary and the secondary carriers (the latter is water). Soap — the classical surface-active agent — is unsuitable because of its susceptibility to Ca and Mg ions. Saponin and similar natural substances have been completely ousted in the agrobiological industry by the cheaper synthetic compounds, such as fatty alcohol sulphates, alkyl-aryl sulphonates, polyethylene glycols, etc. One or two per cent of such substances is sufficient to render a preparation wettable.

The wetting-agent content of wettable powders must be such as to ensure that the liquid spray prepared from the powder and water retains sufficient surface tension to prevent rapid drainage of the pesticide from the plant. Nor should the surface tension be so low that troublesome foaming occurs in the spray tank. Further, certain surface-active agents enhance the efficacy of the active substances, so that such agents should be chosen not only for their physico-chemical but also for their biological properties.

In addition to a wetting agent, a wettable powder usually requires a dispersing agent to counter flocculation, i.e. the aggregation of the finely divided

particles. Flocculation would lead to rapid sedimentation of the suspended material and this would be attended with a reduction in the active surface and with blockage of the spray nozzle.

In certain cases wettable powders must also contain a sticker. The latter prolongs the action time, this being especially useful in the case of poorly adhering fungicides and insecticides that work as stomach poisons (see the article cited in ¹), page 146). A highly tenacious residue is undesirable however if the active compound is very stable and particularly toxic to man and domestic animals.

Classification of pesticide preparations

Above we have made a distinction between sprays, dusts and aerosols, according to the way in which a pesticide preparation is applied to the plants. The first and the third groups can be further subdivided, whereupon we arrive at the following classification:

Sprays	$\left\{ \begin{array}{l} \text{solutions} \\ \text{emulsions} \\ \text{solubilized solutions} \\ \text{suspensions} \end{array} \right.$
Dust preparations	
Aerosols	$\left\{ \begin{array}{l} \text{smoke generators} \\ \text{liquid preparations for} \\ \quad \text{atomization} \\ \text{fumigants} \end{array} \right.$

We shall now deal with the principles whereby these different groups are formulated.

Sprays

Solutions

A "true" solution (in contrast to an emulsion or a suspension) gives the best distribution of a pesticide. The insects, mites, fungi or plants to be exterminated are soaked with the pesticide, and when the solvent has evaporated, are left with a homogeneous layer or "crystal carpet" of the pesticide.

Spraying agents intended for use in solution are on the market in both liquid and powder form. The former need only be made up to the right concentration with water (in a few rare instances with paraffin oil) before use; the powder products have to be dissolved in water. To prevent undesirable reactions with hard water, the preparation frequently contains certain other additives. Beyond this, the formulation of water-soluble pesticides presents no serious problems.

Modern pesticides for the control of insects or mites, such as DDT, BHC, lindane, "Tedion V18" and many others are but slightly soluble in water

and cannot be used in the form of true solutions. If these compounds are to be sprayed with water, procedures which will be described in the section on emulsions below, must be adopted. We may mention nicotine (now little used) as one of the few insecticides sufficiently soluble in water for normal application.

Because of its phenolic structure, DNC (4,6-dinitro-orthocresol) which has been used since 1892, occupies a special place. DNC is an acid with a low water solubility, but salts which dissolve satisfactorily in water can be prepared by replacing the "acid" H-atom by Na, K or NH_4 . These salts are biologically less active than the acid; the ammonium salt is the most effective, since after spraying it decomposes fairly quickly into the more active DNC-acid and NH_3 . DNC and its salts are both insecticidal (ovicidal) and herbicidal. But they are also phytotoxic and thus can be employed to kill aphid eggs on fruit trees only in the winter, i.e. during the dormant period of the trees.

We may mention lime sulphur spray (calcium polysulphide) and certain organic mercury compounds as examples of the few fungicides which are sufficiently soluble in water. Well-known fungicides like copper oxychloride, captan, zineb and many others are sprayed in suspension because of their poor solubility in water (see section on suspensions).

A greater number of herbicides are sufficiently soluble in water. Here we shall confine ourselves to the growth substance herbicides³⁾. Most of these are derivatives of phenoxyacetic acid. The principal derivatives have already been referred to: MCPA (2-methyl, 4-chlorophenoxyacetic acid), 2,4-D (2,4-dichlorophenoxyacetic acid) and 2,4,5-T (2,4,5-trichlorophenoxyacetic acid). They exhibit remarkable selectivity between mono- and dicotyledons, and because of this can be used to destroy dicotyledonous weeds growing among monocotyledonous cereals or grasses, without doing any appreciable harm to the latter. To be sure of complete success certain precautions must be taken in their formulation as will be evident from the following.

MCPA preparations are mostly marketed as a concentrated solution of the Na, K or NH_4 salt in water. Since the salts of bivalent metals are sparingly soluble in water, Ca- and Mg-methylchlorophenoxyacetate may be precipitated if the concentrated solution is diluted with hard water. This can lead to blockage of the sprayer and, furthermore, these acetates have no herbicidal action, so that part of the MCPA is inactivated. It is therefore desirable to add sequestering agents during formulation, i.e. substances which form a complex

with Ca and Mg ions so that they can no longer react with the phenoxyacetate. Synthetic dispersing agents are also used for this purpose. But it is important not to add too much of such agents, otherwise the spraying liquid will wet the plants too thoroughly, whereupon monocotyledons may also suffer injury.

The alkali salts of 2,4-D are relatively poorly soluble in water and are thus marketed in powder form only, being sprayed with water in a concentration of a few tenths of a percent. However, the amine salts of 2,4-D are very readily soluble in water; a 40% solution of 2,4-D in the form of the triethanolamine salt is very stable and will withstand a moderate frost. The mono- and di-ethanolamine salts can also be used, also the mono-, di- and trimethylamine and morpholine salts. The correct pH value is important with these concentrated salt solutions. Such preparations are very easy to make up.

The alkali salts of 2,4,5-T are still less soluble in water than those of 2,4-D and for this reason are not used. The organic amine salts of 2,4,5-T are reasonably soluble, but have not been applied on any large scale, mainly because certain 2,4,5-T esters⁵⁾ have an appreciably better herbicidal action. Since these esters are not sufficiently soluble in water, they must be formulated in a special manner, which will be discussed in the following section.

Emulsions

The insecticidal action of mineral oils has been known for some 200 years, but it was less than a century ago that stable emulsions of petroleum hydrocarbons in water were successfully prepared. Such emulsions are two-phase systems consisting of water in which the oil is dispersed as fine droplets. It was about 80 years ago that the first emulsified preparations came on to the market for use against certain scale insects and red spider.

The shortcomings of these products were many and mainly a consequence of the incompatibility of the emulsifier then used — soap — with hard water, which caused the emulsion to separate into oil and water ("breakdown"). When emulsifiers became available which did not suffer from these disadvantages, e.g. protein substances (such as casein), cellulose derivatives and especially the synthetic detergents, mineral oil emulsions could be prepared which have done excellent service to this day in the control of red spider, especially in citrus fruit farming.

It has likewise been found that if an emulsifier is dissolved in a mineral oil or in a tar oil distillation

⁵⁾ Philips tech. Rev. 18, 335, 1956/57 (No. 11).

product, the solution will spontaneously emulsify in water. This has been applied on a large scale in what are called winter washes for combating aphid eggs. Large quantities of tar oils are employed each year for that purpose, notwithstanding the advantages of DNC (see above).

There are two ways in which an emulsive spray can be made up by the user:

- 1) by diluting a concentrated stock emulsion with water,
- 2) by pouring an emulsifiable concentrate into water.

Stock emulsions. Concentrated emulsions of mineral oils in water are fairly easy to prepare. One common technique is to add the oil phase discontinuously under constant stirring, to the water phase in which an emulsifier (usually a colloid such as ammonium caseinate) has been dissolved, until a very viscous emulsion is obtained. The latter is then adjusted to the right concentration with water. In this way stock emulsions can be manufactured having a mineral oil content of 80% which are still fluid. The emulsifier content can be very low, e.g. 0.5%. The advantage of a low emulsifier content is that the emulsion soon breaks down after spraying; the active oil is left adhering to the crop, while the water drains or evaporates off.

If the oil phase of the emulsion contains another substance, such as, for example, DNC, care must be taken that the emulsion has a low pH value, to preclude migration of the DNC from the oil phase to the water phase. This readily occurs in emulsions stabilized with ammonia, and results in the formation of ammonium dinitrocresolate in the water phase. When the emulsion breaks down, part of the active substance is lost with the water that drains off, especially if the spray liquid has a low surface tension.

Another advantage of stock emulsions is that they can be diluted with either soft or hard water; this is a point in their favour over the emulsifiable concentrates. By introducing certain additives special effects can be achieved: prolonged action time, compatibility with fungicides, insecticides or acaricides of quite different formulation, etc.

Over and against these advantages there are certain drawbacks. In the first place we may mention their susceptibility to changes in temperature and their liability to freeze. However, this can be remedied by choosing an appropriate emulsifier; emulsions have been prepared capable of withstanding both frost and tropical temperatures. Another drawback is the danger of undesirable reactions

with the container. A serious consideration also is that many valuable pesticides cannot be made up into stock emulsions because of their readiness to hydrolyze, e.g. a number of thiophosphoric acid esters (malathion, parathion), chlorinated hydrocarbons with labile Cl atoms (toxaphene, dieldrin, aldrin) and esters of 2,4-D and 2,4,5-T (see above).

Because of these disadvantages the stock emulsions have slowly been ousted by the emulsifiable concentrates.

In two cases, however, they have completely held their ground.

1) "White oil", an emulsion of refined mineral oil, is still very widely used, since its phytotoxicity is less than that of emulsifiable concentrates. Perhaps this can be attributed to an "alleviating action" on the part of the emulsifier: when the emulsion breaks down, the heavier water layer containing the colloid may perhaps prevent the oil from coming into direct contact with the plant.

2) Many strong acidic pesticides are easier to formulate as stock emulsions than as emulsifiable concentrates, e.g. pentachlorophenol; one use of this substance is as a defoliant; it quickly rids potato plants of their foliage (this considerably simplifies lifting and likewise counters any infection with potato blight). Well-formulated pentachlorophenol emulsions rapidly break down after application, as is required for this purpose.

Emulsifiable concentrates. These are one-phase systems which spontaneously emulsify on mixing with water, the oil dispersing as droplets (discontinuous phase) throughout the water (continuous phase). The micro-photos in *fig. 1* show: the emulsion formed from an emulsifiable concentrate; the breaking-down of the emulsion; and the residue. In their simplest form, emulsifiable concentrates consist of two components: an active liquid and, dissolved in the latter, an emulsifier. As we shall see shortly, however, more complex compositions are required in practice.

The molecules of an emulsifier contain a hydrophobic part having an affinity for the oil phase, and a hydrophilic part having an affinity for the aqueous phase. When an emulsifiable concentrate is poured into water the emulsifier molecules orientate themselves; they form links at the oil-water interfaces between the two phases which were not originally miscible. The emulsion then forms at once, and is usually of a dual nature, i.e. microscopic examination reveals a dispersed aqueous phase within the dispersed oil phase (*fig. 1*); this is of no practical significance.

Modern emulsifiers employed in the preparation of emulsifiable concentrates almost always consist of two surface-active substances, one of which forms no ions while the other forms anions at the

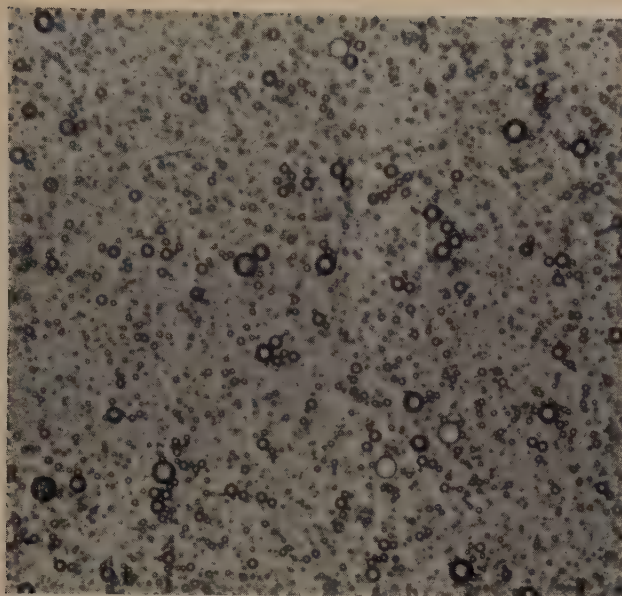
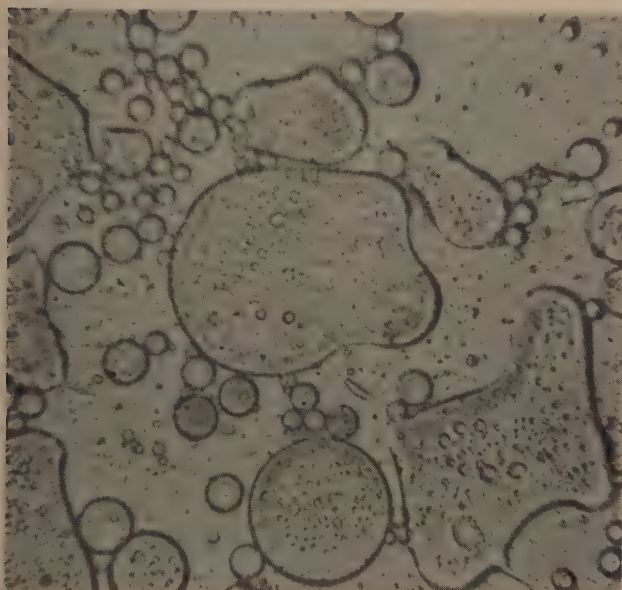


Fig. 1. Photo-micrographs of the acaricide "Tedion V18", formulated as emulsifiable concentrate. Magnification 500 $\times$. *Above*: Emulsion of the emulsifiable concentrate in water. *Centre*: The emulsion is breaking down. Water droplets are emulsified in the oil droplets (still surrounded by water), which are by now fairly large. *Below*: After the emulsion has broken down and the solvent evaporated, the residue is left in the form of fine crystals.



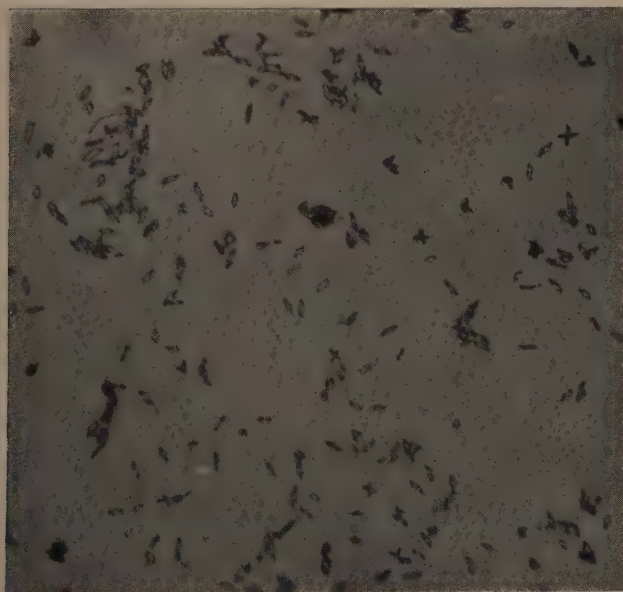
interface. These substances must be chosen individually for each case and must be accurately matched, so that the best possible results are obtained with water of varying hardness. Often a stabilizer is necessary to protect the emulsifier from reaction with decomposition products of the active substance (see below), thus ensuring that the emulsifiability is retained.

The emulsifier is chosen not only for its physical and chemical properties but also for its biological qualities. Some emulsifiers are rather toxic to the plant. When the emulsion has been sprayed, the oil phase that separates out will still contain a fraction of the emulsifier; through its affinity for hydrophobic and hydrophilic interfaces, this emulsifier will penetrate deeper into the plant than the active substance itself (which is usually hydrophobic) and can thus do quite a lot of damage to sensitive crops.

There is another danger to the plant besides the emulsifier; on decomposing, the active substance may give phytotoxic decomposition products. Decomposition can be caused by the presence of too much water or of metals (iron or tin containers). For this reason the water content of emulsifiable concentrates must be kept as low as possible (0.2% is permissible in favourable cases), and metal containers must have a good synthetic-resin lining.

Besides an emulsifier and a stabilizer, an emulsifiable concentrate may require yet another component, namely a solvent for the active substance if this is solid or highly viscous. The solvent is usually a mixture of two or more liquid hydrocarbons (xylene, paraffinic oils).

A widely practised technique, but one that is not to be recommended, is to spray a combination of agro-biological preparations, e.g. an insecticidal emulsion and a fungicidal suspension. The results of such treatment are frequently disappointing, especially if the suspension contains strongly hydrophobic active ingredients. For example, an emulsion of parathion combined with a suspension of zineb shows a strong tendency to flocculate. This soon leads to blockage of the nozzle and likewise reduces the activity of both substances. In general therefore the combinations of emulsions and suspensions should be discouraged.



It is possible to formulate emulsifiable concentrates so that they are physically compatible with other preparations (including suspensions). This entails the mixing of a large amount of strongly hydrophilic emulsifier in the emulsifiable concentrate and it is thus doubtful whether such preparations are an economic proposition.

Solubilized solutions

Solubilized solutions are clear solutions of certain substances in water, being one-phase systems composed of a substance immiscible with water, one or more surface-active agents and certain co-solvents, in the correct proportions. Preparations of this type are characterized by a high emulsifier content (usually predominantly hydrophilic); the co-solvents are usually alcohols. The interfacial tension between the active substance and water is then near enough zero, i.e. there is well-nigh perfect miscibility. The active ingredient in such preparations is particularly finely divided (particles varying from a few tenths to a few thousandths of a micron).

Solubilized DDT preparations have been employed, but have few advantages to offer. On the other hand, solubilized preparations of emphytic agents are important. Naturally both the active substance and the emulsifier must be perfectly innocuous to the plant itself; moreover any breakdown products toxic to man or domestic animals must not accumulate in parts of the plant that are destined for consumption. Here, therefore, there must be a far-reaching investigation into plant and animal toxicity.

The use of the growth substance herbicides in the solubilized state has made little headway. It is to be expected that esters of 2,4-D or 2,4,5-T in the solubilized form will be assimilated by weeds more rapidly than the residue from the usual rapidly breaking-down emulsions. Further research will have to show which formulation is to be preferred.

Suspensions

Suspensions are two-phase systems consisting of finely divided solid particles in a liquid phase (in this instance usually water), see *fig. 2a*. Like emulsions, suspensions can be prepared either by diluting a concentrated paste with water, or by mixing a wettable powder with water.

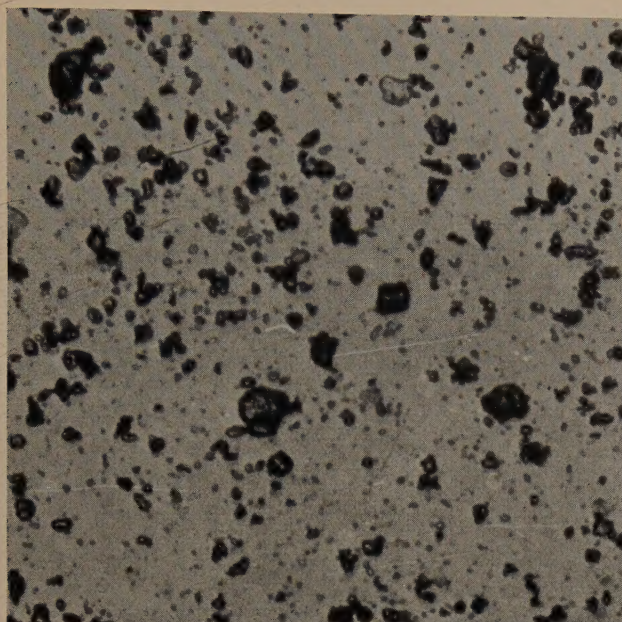
In formulating pastes the difficulty arises that the particles of the active substance must be very small, since otherwise the substance would quickly settle out when suspended in water (according to Stokes' law the rate at which such particles settle out is proportional to the square of their mean

diameter). The grinding of the active substance in colloid mills is not very effective and very costly. Most insecticides and acaricides are not therefore marketed as pastes. The active ingredients for various fungicides can, however, be prepared as a precipitate in an aqueous phase; the particles so obtained are small enough (a few μ or even less) to give suitable pastes without grinding. Very good preparations can be made in this way, e.g. from basic copper salts. A disadvantage of concentrated pastes is that they require rather costly packing. Tins are out of the question because of corrosion; even coated tins are insufficiently reliable, so that all-plastic packing is indicated.

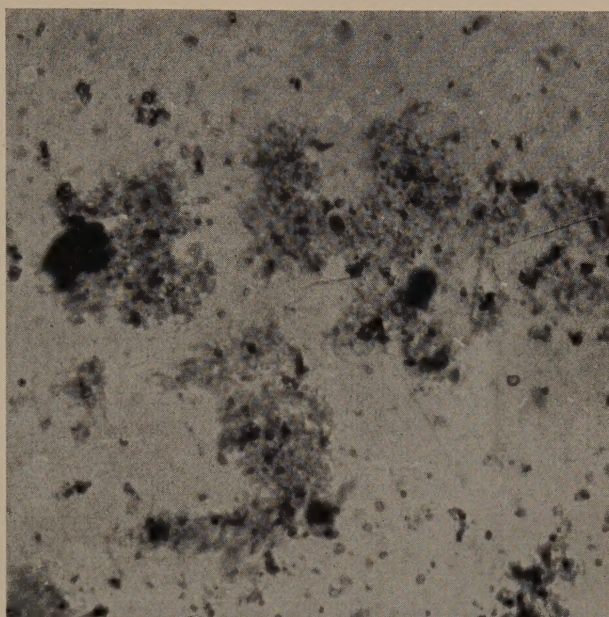
The wettable powders are more practical and cheaper than the pastes. With modern mill equipment, solid active substances together with the accessory substances can be reduced to a mean particle size of 10 μ and even smaller. Modern mills for this purpose, *fig. 3*, have a capacity of 500 to 1000 kg per hour. The contents of active ingredient can be appreciably higher than in pastes, and there are no problems in packing the powder. Nevertheless the formulation of wettable powders calls for special attention. For, in suspension the particles show a tendency to flocculate (*fig. 2b*), especially in the presence of Ca and Mg ions in hard water; the flocculated particles behave like coarser particles, i.e. they sink more quickly. The use of an excess of wetting agents, sequestering agents and dispersing agents leads to some improvement in this respect, but results in poorer adhesion to the plant; and that means little residue, especially in rainy weather.

If a suspension with a good suspensibility is to be obtained, it is essential that the active substance should be well adsorbed by the carrier. Active substances (except for lindane, which can be prepared in a chemically pure state) always contain impurities, these being frequently of an oily nature. Such impurities strongly promote flocculation and complicate the grinding process by greasing up the mills. Such substances as aluminium silicate, synthetic calcium silicate and colloidal silicic acid have been found to make very good carriers. They excel by reason of their fine structure, light weight and their high capacity for adsorbing sticky pesticides. They are fairly expensive, but this disadvantage is to some extent compensated by their strongly hydrophilic nature, which means that a low wetting and dispersing agent content will suffice.

Because the wettable powders have a high content of active substance, freight costs are relatively low. Another advantage is that, by dilution of the wett-



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a

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b

Fig. 2. "Tedion V18" in suspension, *a*) not flocculated, *b*) partly flocculated. Magnification 500 $\times$.

able powder with carriers available in the country of destination, dusting powders, which are dusted dry on the plant (see the following section), can be prepared. At tropical temperatures there is a danger of lumping, especially in the case of BHC and DDT preparations containing 75% or more of the active ingredient; this can be remedied by compounding colloidal silicic acid in the preparation.

Another advantage of wettable powders should be added to those mentioned above, namely that they are in general less toxic to man, domestic animals and plants than the emulsions, and can be more readily combined with other preparations. It is therefore to be expected that emulsifiable concentrates will be used less and less and that the wettable powders will gain ground.

Dust preparations

Dusting agents are dry powders which are ready for use right away. The content of active ingredient varies from a few tenths of a per cent to a few per cent. Owing to the large bulk of inert carrier, it is of great importance that the active ingredient should be fine and evenly distributed throughout the whole mass.

The active ingredient may be solid or fluid. In the former case the dusting powder is usually prepared by mixing a finely ground pre-concentrate of the active ingredient in the carrier. The choice

of the carrier and any other auxiliary substances is mainly determined by the physical properties of the active substance, such as its hardness, melting point, hydrophilic or hydrophobic nature. To ensure that the product does not lose its uniformity during storage and transport or during application, the particles must be of a uniform size; this reduces the effect of large differences in the specific gravities of the various components.

Where the active substance is more or less fluid (nicotine, parathion, malathion, toxaphene, etc), it must usually be applied on to the carrier in the form of an atomized mist (if necessary, having first been diluted with a volatile solvent). This procedure calls for good adsorbing properties on the part of the carrier, especially when pre-concentrates are to be prepared.

The finished product, irrespective of whether it has been prepared by the milling or the atomizing technique, must retain its original consistency. It must not be subject therefore, to aggregation during transport and storage when it may be exposed to pressure and elevated temperature. Aggregation does not usually give any trouble in preparations having a low content of active substance. However, the problems with pre-concentrates containing 80% or more of active substance are numerous. If the active substance has a fairly high vapour pressure (BHC, lindane), the preparation begins to agglomerate markedly at heightened temperature, to give lumps. Auxiliary substances

(conditioners) may be necessary to combat this.

Depending upon the plants that are to be treated, heavy or light dusting powders are used: heavy for low crops, light for trees.

flour, sawdust and tobacco dust. Some carriers are themselves lethal to insects, either extracting water from the insect, or like crude silicic acid, damaging their cuticula by their abrasive action.

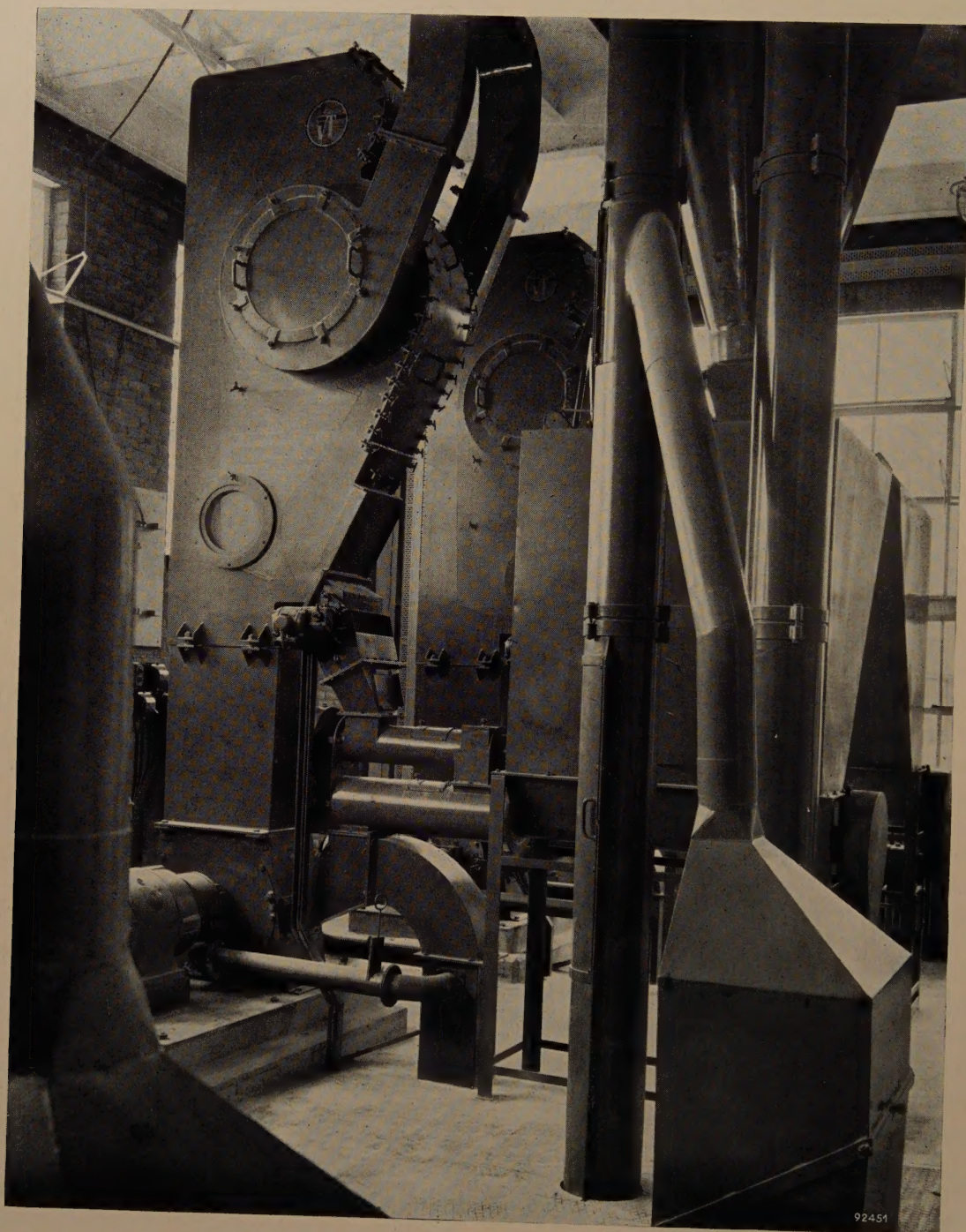


Fig. 3. One of the milling installations in the Amsterdam pesticide factory of N.V. Philips-Roxane.

The most important carriers are: magnesium and aluminium silicate (talc, kaolin), carbonates (dolomite, chalk), materials containing silicic acid (diatomaceous earth), gypsum, sulphur; further, in countries where they can be cheaply obtained: organic materials, such as wheat flour, walnut

It has already been mentioned in the introduction, however, that an abrasive action is bad both for the plant and for the dusting apparatus.

Many pesticides are sensitive to alkalis, thus the carrier should not have an alkaline reaction. Even if it is neutral, a compatibility test should still be

made, since certain impurities bring about catalytic decomposition of the active substance.

Aerosols

Aerosols are systems in which the solid or liquid active substance is extremely finely dispersed throughout a gaseous phase (in the present case, air) and remains suspended in the gas ("micro-mist"). In time the particles become greater by taking up moisture or by aggregation. Sedimentation sets in, so that the active substance settles on the crop. The active substance may work as a respiratory or as a contact poison. In the former case it is most effective in its micro-mist form. The contact action is only satisfactory if the particles are not too small and if the insects are very mobile, e.g. flies, which can take up a fair amount of poison during their flight and die mainly as a result of the contact action.

Aerosols, as has already been remarked, are on the market as fumigation agents, as liquid atomization preparations and as vapour agents.

Smoke generators

Smoke generators are preparations in which the active substance is mixed with an oxidizing agent and with a carrier (usually organic). Once ignited, the mixture gives off a great deal of smoke (fig. 4) consisting of the active substance and reaction products such as water vapour and carbon dioxide. Such preparations are effective only in confined spaces (greenhouses).

The preparation of smoke generators is not without danger, since it involves powerful oxidizing agents such as ammonium nitrate, chlorates, perchlorates or bichromates. It is absolutely essential here that all the components should be mutually compatible, if sudden undesirable reactions are to be avoided. Apart from the choice of oxidizing agents it is also important that the combustion temperature should not be too high; otherwise much of the active substance would be lost by burning. The loss of active substance is nevertheless as much as 20 to 30% even in efficient DDT and lindane smoke-generating preparations.

Liquid preparations for atomization

If a liquid is sprayed at a sufficiently high temperature and pressure a mist is obtained in which the droplets are fine enough to remain suspended in the air. This is another method used for spreading pesticides. The liquid used is a solution of an active substance in an organic solvent. Depending upon the size of the droplets, a blue (very fine) or a white

(coarser) "smoke" is produced. The atomization is often effected nowadays using simply constructed apparatus working with steam. Injurious insects on dense upright crops or in woods can be effectively combated by atomization from aeroplanes. An objection to this practice, however, is that birds

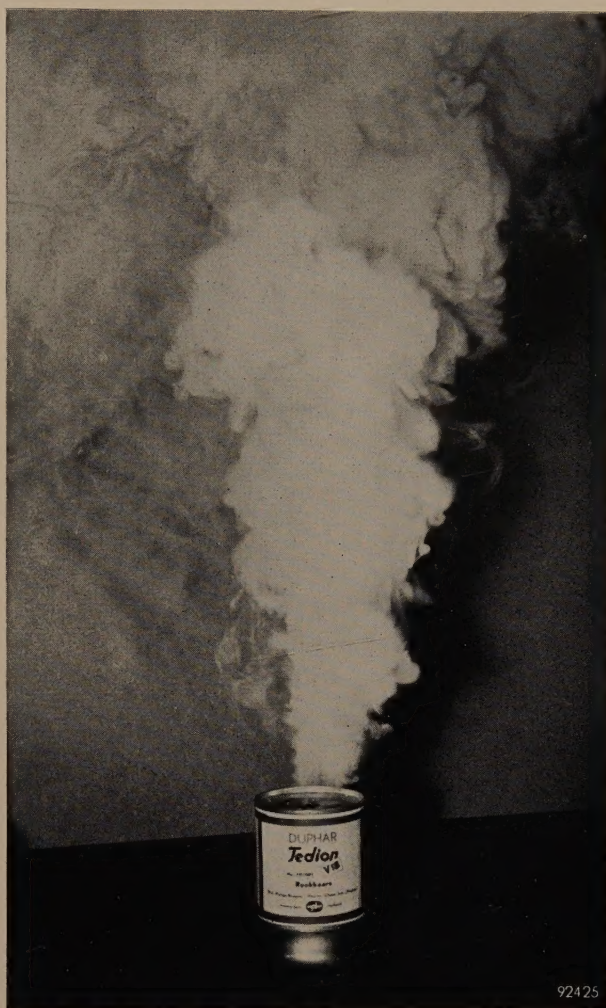


Fig. 4. Smoke evolved by smoke generator.

and useful insects, like bees, frequently suffer much harm. The success of the method, moreover, is highly dependent upon the wind.

The solvent must have a high specific gravity (so that the mist does not settle too slowly); further, it should not burn readily and should have a low phytotoxicity, since the amount of solvent in the mist is quite considerable.

Fumigants

Fumigants are preparations containing an active substance with a high vapour pressure; the active substance is distributed by vaporization which

occurs at relatively low temperatures. The vapour molecules are imagined to combine to form a mist; this is why fumigants are classed as aerosols. Formulation is necessary only where the substance is too volatile or insufficiently volatile for normal use; the required rate of evaporation can be obtained by means of suitable additives.

A well-known vapour agent for domestic use is paradichlorobenzene, for combating moths in wardrobes. Like orthodichlorobenzene, this substance can be of good service as a soil disinfectant for exterminating termites. More widely used is "DD mixture", a mixture of dichloropropane and dichloropropylene. This is a liquid with a boiling point around 100 °C; the highly penetrating vapour is especially active in the soil.

Concluding remarks

The requirements with which the quality of agro-biological preparations must comply are becoming ever more exacting, and this calls for ceaseless work on their formulation. Among the fundamental problems that are engaging the attention of the Philips-Roxane formulation laboratory, are the flocculation phenomena and the stabilization of preparations at high temperature; these problems must be solved for each substance individually.

Naturally these physico-chemical questions must be studied with reference to the biological properties of the preparation. With a view to obtaining the optimum biological activity a great deal of attention is being given to formulations, especially for the pesticides discovered and developed by Philips-Roxane.

There are indications that the biological activity can be very much enhanced by careful choice the auxiliary substances such as carriers and surface-active agents. It is therefore not enough to test a preparation that is under development merely by physico-chemical methods; the development of a preparation can be regarded as complete only when that preparation has been put to exhaustive biological evaluation in the laboratory and in the field, and when the results of such evaluations are in accord with the preparation's physico-chemical properties. As more selective agents, particularly emphytic agents are discovered, the formulation of agro-biological preparations will become still more complex; for good results there will have to be close cooperation between the chemist and the biologist.

Summary. The control of injurious insects, red spiders, fungi and weeds is mainly done by means of chemicals: insecticides, acaricides, fungicides and herbicides, which come under the general heading of pesticides. An agro-biological preparation consists of a pesticide and various auxiliary substances (carrier, wetting agent, sticker, emulsifier, stabilizer, etc.). The compounding or formulation of these preparations is extremely important. According to the way in which the preparation is to be used, we differentiate between sprays, dusts and aerosols. The sprays are subdivided into solutions, emulsions (stock emulsions and emulsifiable concentrates), solubilized solutions and suspensions; the aerosols into smokes, liquid preparations for atomization, and fumigants. The optimum size of the particles of the active substance, the chemical compatibilities of the components of a preparation, phytotoxicity, lumping of powder preparations, flocculation of suspensions, the breaking-down of emulsions, the influence of changes in temperature and of the packing, are some of the questions touched upon in this article and occupying the attention of the "formulation chemist".
